A Novel Synthetic Route for the Synthesis of 4,6-Diaryl-2-methyl-1,3-benzoxazoles

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A new synthetic route was developed for the synthesis of 4,6-diaryl-2-methyl-1,3-benzoxazoles and their hydrogenated derivatives. The target compounds were obtained *via* the Neber rearrangement from 3,5-diaryl-2-cyclohexen-1-ones. The formation of the isomers in the dihydro derivatives was explained by the [1,5] sigmatropic shift of hydrogen under thermal condition. DDQ was employed for the dehydrogenation of the dihydro benzoxazoles.

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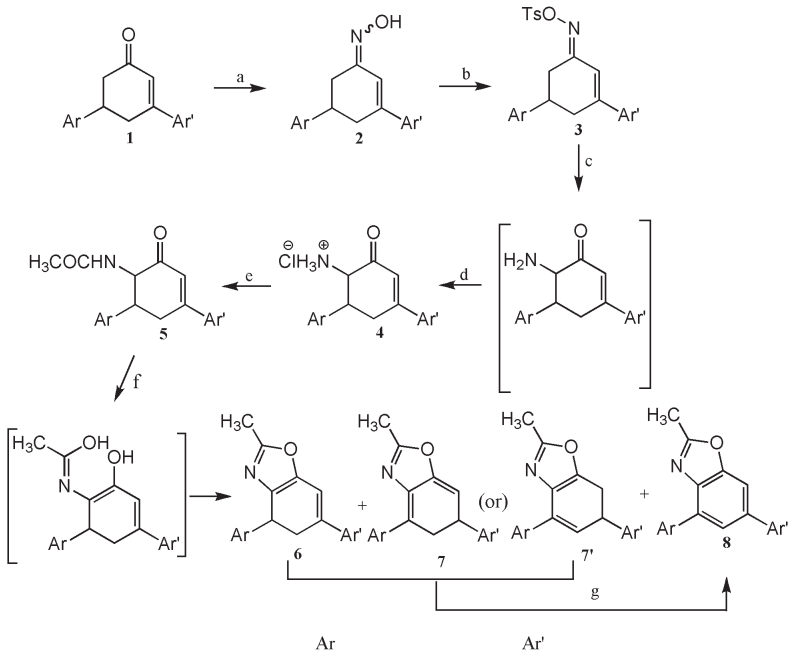
Introduction.

The chemistry and pharmacology of suitably substituted benzoxazoles have attracted the attention of organic chemists and biochemists. A large number of benzoxazoles and their derivatives have been shown to possess interesting biological activities [1-6] and photophysical properties [7,8]. A variety of methods are available to synthesize such benzoxazoles [9-12]. Here we report a new facile route to synthesize 4,6-diaryl-2-methyl-1,3-benzoxazoles employing the Neber rearrangement. The Neber rearrangement, which describes the formation of α-aminoketones by the treatment of sulfonic esters of ketoximes with potassium ethoxide, has been extensively reviewed [13,14]. It is a key step in the synthesis of several aminoacids [15], aminoalcohols [16], leucoanthocyanins [17], and 3-aminopiperidines [18]. The asymmetric synthesis of 2*H*-azirine-2-phosphates involving Neber rearrangement has also been recently reported [19].

Scheme 1



	Ar	Ar'
a	Phenyl	Phenyl
b	Phenyl	4-Chlorophenyl
c	4-Methoxyphenyl	4-Chlorophenyl
d	Phenyl	2-Naphthyl
e	4-Methylphenyl	Phenyl
f	4-Methoxyphenyl	Phenyl
g	2-Furyl	2-Thienyl
h	4-Methylphenyl	2-Thienyl
i	2-Furyl	Phenyl
j	2-Thienyl	4-Methylphenyl

Reagents and conditions: (a) $NH_2OH.HCl$, CH_3COONa, aqueous alcohol, reflux, 85-96%; (b) TsCl, pyridine, 0°C, 86-96%; (c) NaOEt, EtOH, rt, 58-65%; (d) 10% HCl; (e) CH_3COONa, Ac_2O; (f) $POCl_3$, reflux, 65-95%; (g) DDQ, dioxane, 110°C, 82-87%.

The key chiral intermediate for the synthesis of a β_3 agonist was prepared *via* an improved Neber rearrangement [20]. The intermediate 1-aminoindolo[2,3-*a*]quinolizidine for the synthesis of *E*-azaeburamine type compounds has been successfully prepared employing this rearrangement [21].

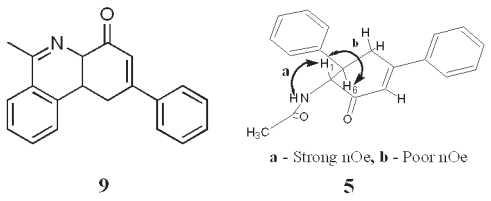
Results and Discussion.

We planned to synthesize the target compounds **8** from 3,5-diarylcyclohex-2-en-1-ones, **1** involving the base- catalyzed Neber rearrangement (Scheme 1). The 3,5-diarylcyclohex-2-en-1-ones **1** were prepared from the corresponding chalcones and ethyl acetoacetate by the reported method [22]. The enones were converted to their oximes **2** by refluxing the alcoholic solution of the enones with an excess of hydroxylamine hydrochloride and sodium acetate. The ^{13}C NMR spectrum of the crude oxime **2** showed the formation of a major amount of *E*-isomer and a trace amount of the *Z*-isomer. In the case of **2a**, the C-2 olefinic carbon appears at 119.9 ppm for the *E*-isomer and at 112.4 ppm for the *Z*-isomer but in the ^1H NMR spectrum, the olefinic hydrogen of the *E*-isomer appears at 6.67 ppm and that of the *Z*-isomer gets deshielded and merged with the other aromatic protons. The major *E*-isomer was isolated by a careful recrystallization of the mixture from ethanol and characterized by NMR. The *E*-oximes **2** were converted to their corresponding tosylates **3** by treating the oximes with a large excess of *p*-toluenesulfonyl chloride in pyridine at 0 °C.

The key step of our synthetic route is the conversion of oxime tosylate **3** to the amine hydrochloride **4** by a Neber rearrangement. When the oxime tosylate **3** was treated with sodium ethoxide in absolute ethanol, it was converted to the corresponding α-amino ketone, which was then converted to amine hydrochloride **4** by treatment with hydrochloric acid.

It should be mentioned that the formation of the azirine intermediate in a Neber rearrangement was independent of the configuration of the oxime tosylate. The crude oxime mixture containing both *E* and *Z* isomers was also converted into the corresponding mixture of oxime tosylates (characterized by NMR) which on treatment with sodium ethoxide was converted to the amine with the same percentage yield as in the case of pure *E*-isomer of the tosylate.

The *N*-(2-oxo-4,6-diaryl-3-cyclohexyl) acetamides **5** have been obtained by acetylating the hydrochlorides of α-amino ketones **4** with acetic anhydride and sodium acetate in good yield. Although we had planned to cyclise the acetates **5** with phosphoryl chloride to get **6**, there is a possibility for the formation of phenanthridone derivative **9** [23] (Figure 1). But the expected product **6** was isolated along with its isomer **7** and the aromatized product **8**. The overall yield of this reaction was excellent and the percentage yields of the hydrogenated products **6**, **7** and the aromatized product **8** were calculated from ^1H NMR (Table 1).



9 **5**

Figure 1

Since the isomers **6** and **7** have the same R_f value under identical conditions, these compounds could not be separated. The aromatized product was separated from the mixture through silica column chromatography. It should be mentioned that the acetates **5g** and **5i**, in which the 2-furyl group is at 5 position, upon treatment with phosphorous oxychloride gave entirely the aromatized product **8g** and **8i** respectively with no trace of **6** or **7**.

Table 1

Yields of **6**,**7** and **8** Obtained from $POCl_3$ Cyclisation of **5**

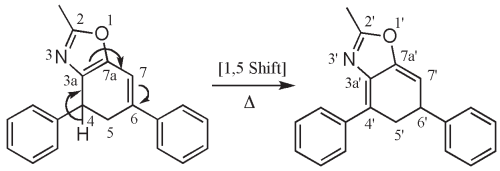
Starting acetate	Overall yield of **6**, **7** and **8** %	Percentage yield* **6**	**7**	**8**
5a	93	77	8	15
5b	90	75	15	10
5c	94	86	3	11
5d	92	31	54	15
5e	95	80	8	12
5f	89	60	30	10
5g	80	0	0	100
5h	91	29	59	12
5i	65	0	0	100
5j	93	39	48	13

* calculated from ^1H NMR based on the 2-methyl signal intensity.

In the case of compound **6a** (Scheme 2), the proton H-7 appeared at 6.75 ppm as a broad singlet as it has two protons in the allylic position. The protons H-5a and H-5b appeared as two different doublets of doublets of doublets (ddd) at 3.10 ppm (J = 17.1, 8.7, 0.9 Hz) and 3.33 ppm (J = 17.1, 9.0, 1.8 Hz) respectively and H-4 appeared as doublet of doublets with coupling constants 9.0 and 8.7 Hz. For compound **7a**, the proton H-7' appeared as a doublet of triplets at 6.26 ppm with coupling constants 3.6 and 1.8 Hz and H-6' appeared as another doublet of triplets with coupling constants 7.2 and 3.6 Hz. The geminal hydrogens H-5'a and H-5'b appeared as a multiplet in between 3.69 ppm and 3.87 ppm. The selected ^1H and ^{13}C NMR signals of **6** and **7** are given in Tables 2 and 3 respectively.

The formation of **7** can be explained by the [1,5] sigmatropic rearrangement (Scheme 2) of **6**, which is a symmetry allowed process under thermal condition. The percentage of **7**, in some cases, is high even though the oxazole ring loses its aromatic sextet. The aromatized product **8**

Scheme 2



6a **7a**

dicyano-1,4-benzoquinone (DDQ) was employed for dehydrogenation in dioxane and the reaction was completed within ten hours. The yields and reaction times of the DDQ dehydrogenation of **6/7** are given in the experimental section. All the benzoxazoles **8** are solids and were fully characterized by 1D and 2D NMR experiments and all the compounds gave satisfactory C, H and N analytical data.

The configuration of the acetate **5** may play a role in the cyclisation step with the possibility for the formation of

Table 2

^{1}H NMR* data of **6** and **7**

Compd.	2-Me s	H-4 dd	H-5a ddd	H-5b ddd	H-7 bs	2'-Me s	H-5'a & 5'b m	H-6' dt	H-7' td	Others
6,7a	2.45	4.28 (*J* = 9.0, 8.7 Hz)	3.10 (*J* = 17.1, 8.7, 0.9 Hz)	3.33 (*J* = 17.1, 9.0, 1.8 Hz)	6.75	2.42	3.69-3.87	4.70 (*J* = 7.2, 3.6 Hz)	6.26 (*J* = 3.6, 1.5 Hz)	7.18-7.45, m
6,7b	2.45	4.27 (*J* = 9.0, 8.7 Hz)	3.04 (*J* = 17.1, 8.7, 0.9 Hz)	3.28 (*J* = 17.1, 9.0, 1.5 Hz)	6.73	2.42	3.65-3.83	4.69 (*J* = 6.9, 3.6 Hz)	6.24 (*J* = 3.6, 1.8 Hz)	7.19-7.39, m
6,7c	2.46	4.23 (*J* = 9.0, 8.7 Hz)	3.02 (*J* = 16.8, 8.7, 0.9 Hz)	3.26 (*J* = 16.8, 9.0, 1.5 Hz)	6.73	2.43	3.70-3.90	4.65 (*J* = 6.9, 3.6 Hz)	6.23 (*J* = 3.6, 1.8 Hz)	3.76, s, 3.80, s 6.82-7.37, m
6,7d	2.45	4.31 (*J* = 9.0, 9.0 Hz)	3.21 (*J* = 17.1, 9.0, 1.2 Hz)	3.42 (*J* = 17.1, 9.0, 1.5 Hz)	6.89	2.43	3.79-3.97	4.73 (*J* = 7.2, 3.6 Hz)	6.41 (*J* = 3.6, 1.5 Hz)	7.20-7.85, m
6,7e	2.41	4.21 (*J* = 9.0, 8.7 Hz)	3.06 (*J* = 17.1, 8.7, 0.9 Hz)	3.27 (*J* = 17.1, 9.0, 1.5 Hz)	6.72	2.39	3.65-3.82	4.64 (*J* = 7.2, 3.6 Hz)	6.24 (*J* = 3.6, 1.8 Hz)	2.27, s, 2.28, s 7.06-7.42
6,7f	2.42	4.21 (*J* = 9.0, 8.7 Hz)	3.05 (*J* = 17.1, 8.7, 1.2 Hz)	3.27 (*J* = 17.1, 9.0, 1.8 Hz)	6.73	2.39	3.67-3.85	4.63 (*J* = 7.2, 3.9 Hz)	6.23 (*J* = 3.9, 1.8 Hz)	3.72, s, 3.73, s 6.80-7.46, m
6,7h	2.43	4.23 (*J* = 9.3, 8.7 Hz)	3.07 (*J* = 17.1, 8.7, 1.2 Hz)	3.28 (*J* = 17.1, 9.3, 1.5 Hz)	6.76	2.41	3.68-3.87	4.63 (*J* = 6.9, 3.6 Hz)	6.31 (*J* = 3.6, 1.5 Hz)	2.29, s, 2.30, s 6.95-7.20, m
6,7j	2.45	4.53 (*J* = 8.7, 8.7 Hz)	3.16 (*J* = 16.8, 8.7, 0.9 Hz)	3.31 (*J* = 16.8, 8.7, 1.8 Hz)	6.69	2.43	3.62-3.78	4.99 (*J* = 7.2, 3.6 Hz)	6.28 (*J* = 3.6, 1.8 Hz)	2.33, s, 2.34, s 6.89-7.54, m

*chemical shifts in ppm and coupling constants in Hz.

was obtained from either **6** or **7**, probably by air oxidation.

Though **7** has been assigned a structure as shown in Scheme 2, it could have an alternate structure **7'** (Scheme 1) with double bonds between 3a/7a and 4/5 in the central ring. This structure **7'**, which may result due to a subsequent [1,5] sigmatropic shift on **7**, also accounts for the available NMR data.

Although the dihydrobenzoxazoles are easily air oxidized, to achieve complete conversion of **6/7** to **8**, 2,3-dichloro-5,6-

Table 3

Selected ^{13}C NMR* Data of **6** and **7**

Compd.	C-4	C-5	C-7	C-5'	C-6'
6,7a	40.0	37.8	111.9	26.5	43.5
6,7b	39.9	37.6	112.2	26.4	43.5
6c	39.1	37.8	112.2	**	**
6,7d	40.1	37.6	112.2	26.5	43.6
6,7e	39.6	37.7	111.8	26.5	43.1
6,7f	39.1	37.8	111.8	26.4	42.6
6,7h	39.5	37.6	110.0	26.7	42.8
6,7j	35.1	37.9	111.0	26.5	37.9

* Chemical shifts in ppm; ** The concentration of **7c** is so small in the mixture to get recognizable signals.

phenanthridone derivative **9** (Figure 1). The coupling constant of protons H-1 and H-6 are very high (around 13 Hz) suggesting that they are perfectly *anti* to each other. This has also been confirmed from the NOESY spectrum. The proton H-1 made very strong nOe contact with the NH proton but it showed a very weak correlation with H-6. The preferable geometry of the acetate **5** is shown in Figure 1. Although this diequatorial like arrangement of the acetamido group and the aryl ring is not unfavorable for cyclisation to yield **9**, the easy enolisation at carbonyl driven by the presence of conjugation makes the other cyclisation to yield **6** easier.

EXPERIMENTAL

All melting points are uncorrected. ^{1}H, ^{13}C and the related 2D NMR spectra were recorded on a Bruker 300 MHz instrument in $CDCl_3$ using TMS as internal standard. Chemical shifts are given in parts per million (δ-scale) while coupling constants are in Hertz. IR spectra were recorded on a Jasco FT-IR instrument (KBr). Elemental analyses were carried out on a Vario EL III instrument. Column chromatography was carried out on silica gel (60-120 mesh) using petroleum ether-ethyl acetate as eluent.

Preparation of 3,5-diaryl-2-cyclohexen-1-one (**1**).

General Procedure.

To a mixture of 1,3-diaryl-2-propen-1-one (20 mmol) and ethyl acetoacetate (2.60 g, 20 mmol) in dry ether (60 mL), a solution of sodium ethoxide (0.276 g, 4 mmol) in ethanol (20 mL) was added. The reaction mixture was stirred for 20 h, acidified with acetic acid (4 mL) and the organic layer was washed with water. The ethereal solution was dried (Na_2SO_4) and evaporated to get the β-ketoester, which was refluxed with 10% sodium hydroxide solution (100 mL) for 6 h. After completion of the reaction, the mixture was extracted with chloroform and dried. The pure product of **1** was obtained either by recrystallization from alcohol or by running through silica column.

3,5-Diphenyl-2-cyclohexen-1-one (**1a**).

This compound was obtained as colorless crystals (alcohol), yield 75%, mp 89-90 ºC (89-90 ºC) [22].

3-(4-Chlorophenyl)-5-phenyl-2-cyclohexen-1-one (**1b**).

This compound was obtained as colorless crystals (alcohol), yield 72%, mp 104-105 ºC; ir (KBr) 3031, 2938, 1654, 1594, 1488, 1402, 1261, 1093, 1014 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.65-2.81 (m, 2 H), 2.84-3.04 (m, 2 H), 3.39-3.50 (m, 1 H), 6.48 (d, *J* = 2.1 Hz, 1 H), 7.25-7.40 (m, 7 H), 7.48 (d, *J* = 8.4 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 36.1, 40.9, 43.8, 125.3, 126.7, 127.1, 127.4, 128.8, 129.0, 136.2, 136.7, 142.9, 157.2, 199.0.

Anal. Calcd for $C_{18}H_{15}ClO$: C, 76.46; H, 5.35%. Found: C, 76.5; H, 5.3%.

3-(4-Chlorophenyl)-5-(4-methoxyphenyl)-2-cyclohexen-1-one (**1c**).

This compound was obtained as colorless crystals (alcohol), yield: 76%, mp 129-130 ºC. ir (KBr) 3048, 2940, 1652, 1590, 1486, 1400, 1270, 1099, 1015 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.62-2.80 (m, 2 H), 2.84-3.02 (m, 2 H), 3.34-3.45 (m, 1 H), 3.81 (s, 3 H), 6.47 (d, *J* = 2.4 Hz, 1 H), 6.91 (d, *J* = 8.7 Hz, 2 H), 7.21 (d, *J* = 8.7 Hz, 2 H), 7.37 (d, *J* = 8.4 Hz, 2 H), 7.48 (d, *J* = 8.4 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 36.9, 40.6, 44.6, 55.7, 114.6, 125.7, 127.9, 128.2, 129.5, 135.6, 136.6, 137.2, 157.8, 159.0, 199.6.

Anal. Calcd for $C_{19}H_{17}ClO_2$: C, 72.96; H, 5.48%. Found: C, 73.0; H, 5.5%.

3-(2-Naphthyl)-5-phenyl-2-cyclohexen-1-one (**1d**).

This compound was obtained as colorless crystals (alcohol), yield: 78%, mp 157-158 ºC; ir (KBr) 3033, 2938, 1650, 1594, 1480, 1404, 1275, 1080, 1021 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.70-2.85 (m, 2 H), 2.96-3.24 (m, 2 H), 3.44-3.53 (m, 1 H), 6.67 (d, *J* = 2.1 Hz, 1 H), 7.28-7.44 (m, 5 H), 7.50-7.53 (m, 2 H), 7.66 (d, *J* = 8.4 Hz, 1 H), 7.82-7.87 (m, 3 H), 8.01 (s, 1 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 36.7, 41.5, 44.4, 123.7, 125.8, 126.8, 127.2, 127.3, 127.6, 127.8, 128.1, 129.0, 129.1, 129.3, 133.5, 134.4, 135.8, 143.7, 158.8, 199.7.

Anal. Calcd for $C_{22}H_{18}O$: C, 88.56; H, 6.08%. Found: C, 88.6; H, 6.2%.

5-(4-Methylphenyl)-3-phenyl-2-cyclohexen-1-one (**1e**).

This compound was obtained as colorless crystals (alcohol), yield 74%, mp 105-106 ºC (106.5-107.5 ºC) [22].

5-(4-Methoxyphenyl)-3-phenyl-2-cyclohexen-1-one (**1f**).

This compound was obtained as colorless crystals (alcohol), yield: 70%, mp 106-107 ºC (107-108 ºC) [22].

5-(2-Furyl)-3-(2-thienyl)-2-cyclohexen-1-one (**1g**).

This compound was obtained as colorless crystals (alcohol), yield 69%, mp 87-88 ºC; ir (KBr) 3052, 2911, 1650, 1592, 1417, 1319, 1257, 1090, 1010 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.63-2.99 (m, 3 H), 3.21 (dd, *J* = 17.7, 4.5 Hz, 1 H), 3.53-3.61 (m, 1 H), 6.12 (d, *J* = 3.0 Hz, 1 H), 6.33 (dd, *J* = 3.0, 1.8 Hz, 1 H), 6.48 (d, *J* = 2.1 Hz, 1 H), 7.12 (dd, *J* = 4.5, 4.2 Hz, 1 H), 7.37-7.47 (m, 3 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 33.3, 34.3, 41.8, 105.3, 110.6, 122.8, 128.2, 128.8, 129.6, 142.1, 142.7, 151.2, 156.5, 198.2.

Anal. Calcd for $C_{14}H_{12}O_2S$: C, 68.83; H, 4.95%. Found: C, 68.9; H, 4.9%.

5-(4-Methylphenyl)-3-(2-thienyl)-2-cyclohexen-1-one (**1h**).

This compound was obtained as colorless crystals (alcohol), yield: 70%, mp 116-117 ºC; ir (KBr): 3043, 2915, 1655, 1594, 1470, 1310, 1255, 1094, 1016 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.35 (s, 3 H), 2.64-2.70 (m, 2 H), 2.84 (ddd, *J* = 17.4, 11.1, 2.1 Hz, 1 H), 3.07 (dd, *J* = 17.4, 4.2 Hz, 1 H), 3.33-3.43 (m, 1 H), 6.50 (d, *J* = 2.1 Hz, 1 H), 7.07 (dd, *J* = 5.1, 3.9 Hz, 1 H), 7.17-7.19 (m, 4 H), 7.36 (d, *J* = 3.9 Hz, 1 H), 7.42 (dd, *J* = 5.1, 0.6 Hz, 1 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 21.5, 36.6, 40.7, 44.6, 122.8, 127.1, 128.1, 128.8, 129.5, 130.0, 137.2, 140.5, 142.8, 152.0, 199.2.

Anal. Calcd for $C_{17}H_{16}OS$: C, 76.08; H, 6.01%. Found: C, 76.2; H, 5.9%.

5-(2-Furyl)-3-phenyl-2-cyclohexen-1-one (**1i**).

This compound was obtained as colorless crystals (alcohol), yield: 73%, mp 44-45 ºC; ir (KBr) 3030, 2937, 1651, 1596, 1488, 1405, 1261, 1094, 1010 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.68 (dd, *J* = 16.5, 11.4 Hz, 1 H), 2.84 (dd, *J* = 16.5, 4.2 Hz, 1 H), 2.96 (ddd, *J* = 17.7, 9.9, 2.1 Hz, 1 H), 3.16 (dd, *J* = 17.7, 4.8 Hz, 1 H), 3.52-3.62 (m, 1 H), 6.11 (m, 1 H), 6.32 (dd, *J* = 3.0, 1.8 Hz, 1 H), 6.47 (d, *J* = 1.8Hz, 1 H), 7.36 (dd, *J* = 1.8, 0.9 Hz, 1 H), 7.40-7.43 (m, 3 H), 7.54-7.58 (m, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 33.6, 34.7, 41.8, 105.2, 110.6, 125.6, 126.6, 129.3, 130.6, 138.7, 142.0, 156.7, 158.5, 198.8.

Anal. Calcd for $C_{16}H_{14}O_2$: C, 80.65; H, 5.92%. Found: C, 80.7; H, 6.0%.

3-(4-Methylphenyl)-5-(2-thienyl)-2-cyclohexen-1-one (**1j**).

This compound was obtained as colorless crystals (alcohol), yield 74%, mp 72-73 ºC; ir (KBr) 3073, 2927, 1650, 1596, 1413, 1359, 1268, 1085, 1015 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.37 (s, 3 H), 2.68 (dd, *J* = 16.5, 11.4 Hz, 1 H), 2.86-2.97 (m, 2 H), 3.18 (dd, *J* = 17.7, 4.2 Hz, 1 H), 3.67-3.74 (m, 1 H), 6.49 (d, *J* = 2.1 Hz, 1 H), 6.92 (m, 1 H), 6.97 (dd, *J* = 5.1, 3.3 Hz, 1 H), 7.18-7.24 (m, 3 H), 7.45 (d, *J* = 8.4 H, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 21.8, 36.7, 37.3, 45.4, 123.9, 124.0, 124.9, 126.6, 127.3, 130.0, 135.6, 141.2, 147.7, 158.6, 198.23.

Anal. Calcd for $C_{17}H_{16}OS$: C, 76.08; H, 6.01%. Found: C, 76.1; H, 6.0%.

Preparation of 3,5-Diaryl-2-cyclohexen-1-one Oxime (**2**).

General Procedure.

To a solution of 3,5-diaryl-2-cyclohexen-1-one **1** (3.0 g) in minimum amount of alcohol, a saturated solution of 6 g of hydroxylamine hydrochloride and 9 g of sodium acetate was

added. The reaction mixture was refluxed for an hour on a water bath and poured into crushed ice, filtered and dried. The major *E* isomer was crystallized out in alcohol from the crude mixture of *E* and *Z* isomers.

(*E*)-3,5-Diphenyl-2-cyclohexen-1-one Oxime (**2a**).

This compound was obtained as colorless solid (alcohol), yield 91%, mp 165-66 ºC (167-170 ºC) [24].

(*E*) 3-(4-Chlorophenyl)-5-phenyl-2-cyclohexen-1-one Oxime (**2b**).

This compound was obtained as colorless solid (alcohol), yield 93%, mp 154-155 ºC; ir (KBr) 3260, 3030, 2890, 1605, 1515, 1430, 1370, 1320, 980 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.44 (dd, *J* = 16.8, 12.9 Hz, 1 H), 2.65-2.94 (m, 2 H), 3.07-3.23 (m, 1 H), 3.41 (dd, *J* = 17.1, 3.9 Hz, 1 H), 6.64 (s, 1 H), 7.24-7.49 (m, 9 H), 9.30 (bs, 1 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 29.3, 36.0, 39.7, 121.1, 127.1, 127.3, 127.4, 127.6, 129.1, 134.5, 138.7, 144.1, 144.7, 157.8.

Anal. Calcd for $C_{18}H_{16}ClNO$: C, 72.60; H, 5.42; N, 4.70%. Found: C, 72.7; H, 5.5; N, 4.7%.

(*E*) 3-(4-Chlorophenyl)-5-(4-methoxyphenyl)-2-cyclohexen-1-one Oxime (**2c**).

This compound was obtained as colorless solid (alcohol), yield 85%, mp 188-189 ºC; ir (KBr) 3265, 3045, 2892, 1601, 1498, 1433, 1372, 1325, 975 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.40 (dd, *J* = 16.8, 12.9 Hz, 1 H), 2.61-2.86 (m, 2 H), 3.04-3.14 (m, 1 H), 3.37 (dd, *J* = 17.1, 4.2 Hz, 1 H), 3.82 (s, 3 H), 6.61 (d, *J* = 1.5 Hz, 1 H), 6.90 (d, *J* = 8.7 Hz, 2 H), 7.24 (d, *J* = 8.7 Hz, 2 H), 7.32 (d, *J* = 8.7 Hz, 2 H), 7.42 (d, *J* = 8.7 Hz, 2 H), 8.50 (bs, 1 H).

Anal. Calcd for $C_{19}H_{18}ClNO_2$: C, 69.62; H, 5.53; N, 4.27%. Found: C, 69.7; H, 5.5; N, 4.3%.

(*E*) 3-(2-Naphthyl)-5-phenyl-2-cyclohexen-1-one Oxime (**2d**).

This compound was obtained as colorless solid (alcohol), yield 96%, mp 180-181 ºC; ir (KBr) 3262, 3052, 2896, 1600, 1496, 1427, 1373, 1317, 968 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.50 (dd, *J* = 16.8, 12.9 Hz, 1 H), 2.76-3.09 (m, 2 H), 3.15-3.24 (m, 1 H), 3.45 (dd, *J* = 16.2, 3.6 Hz, 1 H), 6.83 (s, 1 H), 7.29-7.98 (m, 12 H), 9.50 (bs, 1 H).

Anal. Calcd for $C_{22}H_{19}NO$: C, 84.31; H, 6.11; N, 4.47%. Found: C, 84.4; H, 6.2; N, 4.4%.

(*E*) 5-(4-Methylphenyl)-3-phenyl-2-cyclohexen-1-one Oxime (**2e**).

This compound was obtained as colorless solid (alcohol), yield 93%, mp 162-163 ºC; ir (KBr) 3266, 3043, 2895, 1608, 1510, 1425, 1375, 1319, 976 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.35 (s, 3 H), 2.43 (dd, *J* = 17.1, 12.9 Hz, 1 H), 2.68-2.91 (m, 2 H), 3.04-3.17 (m, 1 H), 3.40 (dd, *J* = 16.8, 4.2 Hz, 1 H), 6.66 (s, 1 H), 7.15-7.36 (m, 7 H), 7.49 (d, *J* = 8.1 Hz, 2 H), 9.80 (bs, 1 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 21.5, 29.4, 36.3, 39.3, 120.8, 125.8, 127.3, 128.7, 129.0, 129.8, 136.8, 140.3, 141.9, 145.4, 158.1.

Anal. Calcd for $C_{19}H_{19}NO$: C, 82.28; H, 6.90; N, 5.05%. Found: C, 82.2; H, 6.9; N, 5.0%.

(*E*) 5-(4-Methoxyphenyl)-3-phenyl-2-cyclohexen-1-one Oxime (**2f**).

This compound was obtained as colorless solid (alcohol), yield 88%, mp 164-165 ºC; ir (KBr) 3275, 3055, 2900, 1602, 1497, 1430, 1370, 1326, 960 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.41 (dd, *J* = 16.8, 12.9 Hz, 1 H), 2.62-2.91 (m, 2 H), 3.04-3.16 (m, 1 H), 3.38 (dd, *J* = 17.1, 3.6 Hz, 1 H), 3.81 (s, 3 H), 6.65 (d, *J* = 1.2 Hz, 1 H), 6.89 (d, *J* = 8.7 Hz, 2 H), 7.22-7.37 (m, 5 H), 7.49 (dd, *J* = 8.4, 1.5 Hz, 2 H), 9.20 (bs, 1 H).

Anal. Calcd for $C_{19}H_{19}NO_2$: C, 77.79; H, 6.53; N, 4.77%. Found: C, 77.8; H, 6.6; N, 4.7%.

(*E*) 5-(2-Furyl)-3-(2-thienyl)-2-cyclohexen-1-one Oxime (**2g**).

This compound was obtained as colorless solid (alcohol), yield 89%, mp 134-135 ºC; ir (KBr): 3270, 3045, 2905, 1600, 1495, 1436, 1370, 1326, 1175, 1140, 970 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.55 (dd, *J* = 16.5, 11.4 Hz, 1 H), 2.65-3.06 (m, 2 H), 3.23-3.32 (m, 1 H) 3.38 (dd, *J* = 16.8, 4.2 Hz, 1 H), 6.12 (d, *J* = 2.1 Hz, 1 H), 6.31 (bs, 1 H), 6.66 (bs, 1 H), 7.00-7.35 (m, 4 H), 10.10 (bs, 1 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 26.9, 32.7, 33.0, 104.9, 110.6, 119.0, 124.8, 126.2, 128.2, 138.1, 141.8, 144.6, 156.6, 157.7.

Anal. Calcd for $C_{14}H_{13}NO_2S$: C, 64.84; H, 5.05; N, 5.40%. Found: C, 64.8; H, 5.1; N, 5.3%.

(*E*) 5-(4-Methylphenyl)-3-(2-thienyl)-2-cyclohexen-1-one Oxime (**2h**).

This compound was obtained as colorless solid (alcohol), yield 92%, mp 155-156 ºC; ir (KBr): 3265, 3025, 2898, 1600, 1513, 1427, 1375, 1320, 983 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.35 (s, 3 H) 2.41 (dd, *J* = 17.1, 12.9 Hz, 1 H), 2.61-2.95 (m, 2 H), 3.05-3.16 (m, 1 H), 3.39 (dd, *J* = 16.8, 4.2 Hz, 1 H), 6.66 (s, 1 H), 6.98-7.34 (m, 7 H), 9.20 (bs, 1 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 21.5, 29.4, 36.3, 39.0, 119.0, 124.6, 126.1, 127.2, 127.3, 129.8, 136.9, 139.1, 141.7, 144.8, 157.8.

Anal. Calcd for $C_{17}H_{17}NOS$: C, 72.05; H, 6.05; N, 4.94%. Found: C, 72.0; H, 6.1; N, 5.0%.

5-(2-Furyl)-3-phenyl-2-cyclohexen-1-one Oxime (**2i**).

This compound was obtained as colorless solid (alcohol), yield 86%, mp 148-149 ºC; ir (KBr): 3270, 3056, 2904, 1602, 1496, 1436, 1369, 1326, 1180, 1145, 966 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.57 (dd, *J* = 16.8, 11.4 Hz, 1 H), 2.77 (ddd, *J* = 17.1, 9.9, 1.5 Hz, 1 H), 3.01 (dd, *J* = 17.1, 4.2 Hz, 1 H), 3.22-3.32 (m, 1 H), 3.40 (dd, *J* = 16.8, 4.2 Hz, 1 H), 6.20 (dd, *J* = 3.0, 0.6 Hz, 1 H), 6.31 (dd, *J* = 3.3, 3.0 Hz, 1 H), 6.65 (d, *J* = 1.5 Hz, 1 H), 7.30-7.38 (m, 4 H), 7.50 (dd, *J* = 7.8, 1.2 Hz, 2 H), 9.80 (bs, 1 H); ^{13}C nmr (75 MHz, $CDCl_3$): δ = 26.9, 32.9, 33.0, 104.81, 110.6, 120.8, 125.9, 128.8, 129.0, 140.3, 141.8, 144.5, 157.0, 157.9.

Anal. Calcd for $C_{16}H_{15}NO_2$: C, 75.87; H, 5.97; N, 5.53%. Found: C, 75.8; H, 5.9; N, 5.4%.

(*E*) 3-(4-Methylphenyl)-5-(2-thienyl)-2-cyclohexen-1-one Oxime (**2j**).

This compound was obtained as colorless solid (alcohol), yield 90%, mp 66-67 ºC; ir (KBr): 3266, 3052, 2895, 1599, 1492, 1430, 1370, 1320, 970 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.34 (s, 3 H), 2.51 (dd, *J* = 16.2, 11.4 Hz, 1 H), 2.69-3.04 (m, 2 H), 3.38-3.52 (m, 2 H), 6.64 (d, *J* = 1.8 Hz, 1 H), 6.92-6.96 (m, 2 H), 7.13-7.18 (m, 3 H), 7.39 (d, *J* = 8.1 Hz, 2 H), 9.80 (bs, 1 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 21.6, 30.5, 35.0, 36.9, 120.2, 123.7, 125.8, 126.3, 127.2, 129.7, 137.3, 138.8, 144.6, 149.0, 157.3.

Anal. Calcd for $C_{17}H_{17}NOS$: C, 72.05; H, 6.05; N, 4.94%. Found: C, 72.2; H, 6.0; N, 5.0%.

Preparation of 3,5-diaryl-2-cyclohexen-1-one Oxime Tosylate (**3**).

General Procedure.

To a cold solution of *E*-3,5-diaryl-2-cyclohexen-1-one oxime **2** (8 mmol) in dry pyridine (28 mL), a solution of *p*-toluenesulfonyl chloride (6.0 g, 32 mmol) in dry pyridine (14 mL) was added slowly, maintaining the temperature at 0-2 °C. The stirring was continued at 0 ºC for 2 h and the reaction mixture was poured into crushed ice with vigorous stirring. The product was filtered, washed with water, dried and recrystallised from alcohol.

(*E*)-3,5-Diphenyl-2-cyclohexen-1-one Oxime Tosylate (**3a**).

This compound was obtained as colorless solid (alcohol), yield 95%, mp 136-137 ºC; ir (KBr) 3048, 2923, 2854, 1610, 1494, 1373, 1189, 1180, 1095 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.45 (s, 3 H), 2.52 (dd, *J* = 17.1, 12.9 Hz, 1 H), 2.74 (ddd, *J* = 17.1, 11.1, 2.4 Hz, 1 H), 2.92 (dd, *J* = 17.1, 3.9 Hz, 1 H), 3.05-3.17 (m, 1 H), 3.36 (dd, *J* = 17.1, 3.9 Hz, 1 H), 6.60 (d, *J* = 2.4 Hz, 1 H), 7.23-7.54 (m, 12 H), 7.89 (d, *J* = 8.4 Hz, 2 H).

Anal. Calcd for $C_{25}H_{23}NO_3S$: C, 71.92; H, 5.55; N, 3.35%. Found: C, 71.8; H, 5.5; N, 3.4%.

(*E*) 3-(4-Chlorophenyl)-5-phenyl-2-cyclohexen-1-one Oxime Tosylate (**3b**).

This compound was obtained as colorless solid (alcohol), yield 92%, mp 85-86 ºC; ir (KBr) 3042, 2930, 2845, 1615, 1490, 1375, 1180, 1173, 1098 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.44 (s, 3 H), 2.50 (dd, *J* = 17.4, 13.2 Hz, 1 H), 2.71 (ddd, *J* = 17.4, 11.4, 2.4 Hz, 1 H), 2.86 (dd, *J* = 17.4, 4.2 Hz, 1 H), 3.04-3.20 (m, 1 H), 3.35 (dd, *J* = 17.4, 4.2 Hz, 1 H), 6.57 (d, *J* = 2.4 Hz, 1 H), 7.24-7.40 (m, 11 H), 7.88 (d, *J* = 8.1 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 21.7, 30.3, 35.3, 39.0, 118.4, 126.8, 126.9, 127.3, 128.8*, 128.9, 129.6, 132.6, 135.1, 137.4, 142.9, 145.1, 149.1, 163.9 (*two carbon signals are merged together).

Anal. Calcd for $C_{25}H_{22}ClNO_3S$: C, 66.44; H, 4.91; N, 3.10%. Found: C, 66.4; H, 4.8; N, 3.1%.

(*E*) 3-(4-Chlorophenyl)-5-(4-methoxyphenyl)-2-cyclohexen-1-one Oxime Tosylate (**3c**).

This compound was obtained as colorless solid (alcohol), yield 94%, mp 89-90 ºC; ir (KBr) 3035, 2927, 2840, 1612, 1513, 1371, 1191, 1178, 1093 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.44 (s, 3 H), 2.47 (dd, *J* = 17.1, 13.2 Hz, 1 H), 2.66 (ddd, *J* = 17.1, 11.4, 2.4 Hz, 1 H), 2.83 (dd, *J* = 17.1, 4.2 Hz, 1 H), 2.99-3.10 (m, 1 H), 3.32 (dd, *J* = 17.1, 3.9 Hz, 1 H), 3.81 (s, 3 H), 6.56 (d, *J* = 2.4 Hz, 1 H), 6.89 (d, *J* = 8.7 Hz, 2 H), 7.16 (d, *J* = 8.7 Hz, 2 H), 7.30-7.39 (m, 6 H), 7.89 (d, *J* = 8.4 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 21.7, 30.5, 35.6, 38.2, 55.3, 114.2, 118.4, 126.9, 127.8, 128.9*, 129.6, 132.6, 135.0, 135.1, 137.4, 145.0, 149.2, 158.7, 164.1 (*two carbon signals are merged together).

Anal. Calcd for $C_{26}H_{24}ClNO_4S$: C, 64.79; H, 5.02; N, 2.91%. Found: C, 64.7%; H, 4.9; N, 3.0%.

(*E*) 3-(2-Naphthyl)-5-phenyl-2-cyclohexen-1-one Oxime Tosylate (**3d**).

This compound was obtained as colorless solid (alcohol), yield 96%, mp 164-165 ºC; ir (KBr) 3045, 2928, 2860, 1605, 1498, 1365, 1180, 1175, 1080 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.45 (s, 3 H), 2.55 (dd, *J* = 17.1, 12.9 Hz, 1 H), 2.84 (ddd, *J* = 17.1, 11.4, 2.4 Hz, 1 H), 3.06 (dd, *J* = 17.4, 3.9 Hz, 1 H), 3.13-3.22 (m, 1 H), 3.39 (dd, *J* = 17.4, 3.9 Hz, 1 H), 6.76 (d, *J* = 2.4 Hz, 1 H), 7.28-7.81 (m, 14 H), 7.91 (d, *J* = 8.4 Hz, 2 H).

Anal. Calcd for $C_{29}H_{25}NO_3S$: C, 74.49; H, 5.39; N, 3.00%. Found: C, 74.6; H, 5.5; N, 3.1%.

(*E*) 5-(4-Methylphenyl)-3-phenyl-2-cyclohexen-1-one Oxime Tosylate (**3e**).

This compound was obtained as colorless solid (alcohol), yield 88%, mp 70-71 ºC; ir (KBr) 3050, 2935, 2850, 1613, 1494, 1373, 1188, 1179, 1094 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.34 (s, 3 H), 2.44 (s, 3 H), 2.49 (dd, *J* = 17.4, 13.2 Hz, 1 H), 2.72 (ddd, *J* = 17.4, 11.1, 2.1 Hz, 1 H), 2.89 (dd, *J* = 17.4, 4.2 Hz, 1 H), 3.01-3.13 (m, 1 H), 3.33 (dd, *J* = 17.4, 4.2 Hz, 1 H), 6.59 (d, *J* = 2.1 Hz, 1 H), 7.13-7.15 (m, 4 H), 7.33-7.47 (m, 7 H), 7.89 (d, *J* = 8.1 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 21.0, 21.7, 30.4, 35.6, 38.7, 118.0, 125.4, 126.7, 128.6, 128.7, 129.2, 129.5, 129.6, 132.7, 136.8, 139.0, 140.1, 145.0, 150.6, 164.3.

Anal. Calcd for $C_{26}H_{25}NO_3S$: C, 72.36; H, 5.84; N, 3.25%. Found: C, 72.4; H, 5.8; N, 3.2%.

(*E*) 5-(4-Methoxyphenyl)-3-phenyl-2-cyclohexen-1-one Oxime Tosylate (**3f**).

This compound was obtained as colorless solid (alcohol), yield 86%, mp 119-120 ºC; ir (KBr) 3046, 2925, 2858, 1610, 1489, 1368, 1185, 1176, 1090 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.44 (s, 3 H), 2.47 (dd, *J* = 17.1, 10.2 Hz, 1 H), 2.69 (ddd, *J* = 17.1, 11.1, 2.1 Hz, 1 H), 2.88 (dd, *J* = 17.1, 3.9 Hz, 1 H), 2.96-3.10 (m, 1 H), 3.31 (dd, *J* = 17.1, 3.9 Hz, 1 H), 3.81 (s, 3 H), 6.58 (d, *J* = 2.1 Hz, 1 H), 6.88 (d, *J* = 8.7 Hz, 2 H), 7.17 (d, *J* = 8.7 Hz, 2 H), 7.33-7.45 (m, 7 H), 7.89 (d, *J* = 8.4 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 21.7, 30.6, 35.7, 38.2, 55.3, 114.1, 117.9, 125.6, 127.8, 128.7, 128.8, 129.2, 129.6, 132.7, 135.2, 139.0, 145.0, 150.6, 158.6, 164.3.

Anal. Calcd for $C_{26}H_{25}NO_4S$: C, 69.78; H, 5.63; N, 3.13%. Found: C, 69.9; H, 5.6; N, 3.2%.

(*E*) 5-(2-Furyl)-3-(2-thienyl)-2-cyclohexen-1-one Oxime Tosylate (**3g**).

This compound was obtained as colorless solid (alcohol), yield 90%, mp 82-83 ºC; ir (KBr) 3042, 2930, 2856, 1616, 1490, 1370, 1178, 1170, 1085 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.44 (s, 3 H), 2.53 (dd, *J* = 17.4, 11.4 Hz, 1 H), 2.72 (ddd, *J* = 17.4, 10.2, 1.8 Hz, 1 H), 3.03 (dd, *J* = 17.4, 4.2 Hz, 1 H), 3.17-3.25 (m, 1 H), 3.28 (dd, *J* = 17.1, 4.2 Hz, 1 H), 6.06 (d, *J* = 3.0 Hz, 1 H), 6.30 (dd, *J* = 3.3, 3.0 Hz, 1 H), 6.54 (d, *J* = 1.8 Hz, 1 H), 7.00-7.08 (m, 2 H), 7.22-7.47 (m, 4 H), 7.89 (d, *J* = 8.7 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 22.1, 28.3, 32.4, 32.7, 105.3, 110.6, 116.2, 126.2, 127.7, 128.4, 129.3, 130.1, 133.0, 142.1, 143.2, 143.5, 145.5, 156.3, 163.5.

Anal. Calcd for $C_{21}H_{19}NO_4S_2$: C, 61.00; H, 4.63; N, 3.39%. Found: C, 61.1; H, 4.7; N, 3.5%.

(*E*) 5-(4-Methylphenyl)-3-(2-thienyl)-2-cyclohexen-1-one Oxime Tosylate (**3h**).

This compound was obtained as colorless crystals (alcohol), yield 89%, mp 143-144 ºC; ir (KBr) 3045, 2928, 2850, 1610, 1490, 1373, 1187, 1179, 1093 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.35 (s, 3 H), 2.44 (s, 3 H), 2.45 (dd, *J* = 17.1, 13.2 Hz, 1 H), 2.68 (ddd, *J* = 17.1, 12.3, 1.8 Hz, 1 H), 2.92 (dd, *J* = 17.1, 3.9 Hz, 1 H), 2.99-3.09 (m, 1 H), 3.29 (dd, *J* = 17.1, 3.9 Hz, 1 H), 6.57 (d, *J* = 1.8 Hz, 1 H), 7.00 (t, *J* = 3.9 Hz, 1 H), 7.12-7.46 (m, 8 H), 7.89 (d, *J* = 8.4 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 21.5, 22.2, 30.9, 35.9, 38.7, 116.2, 127.0, 127.2, 127.6, 128.4, 129.3, 129.9, 130.1, 133.1, 137.3, 140.3, 143.6, 144.3, 145.4, 164.6.

Anal. Calcd for $C_{24}H_{23}NO_3S_2$: C, 65.88; H, 5.30; N, 3.20%. Found: C, 65.8; H, 5.2; N, 3.1%.

5-(2-Furyl)-3-phenyl-2-cyclohexen-1-one Oxime Tosylate (**3i**).

This compound was obtained as colorless crystals (alcohol), yield 91%, mp 128-129 ºC; ir (KBr) 3048, 2925, 2854, 1616, 1485, 1365, 1189, 1182, 1094 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.44 (s, 3 H), 2.63 (dd, *J* = 17.1, 10.8 Hz, 1 H), 2.76 (ddd, *J* = 17.1, 9.6, 1.8 Hz, 1 H), 2.99 (dd, *J* = 17.1 4.2 Hz, 1 H), 3.18-3.28 (m, 1 H), 3.32 (dd, *J* = 17.1, 4.2 Hz, 1 H), 6.06 (d, *J* = 3.0 Hz, 1 H), 6.29 (t, *J* = 3.0 Hz, 1 H), 6.55 (d, *J* = 1.8 Hz, 1 H), 7.34-7.46 (m, 8 H), 7.90 (d, *J* = 8.1 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 22.1, 28.3, 32.7, 32.8, 105.3, 110.6, 118.5, 126.0, 129.1, 129.2, 129.7, 130.1, 133.1, 139.3, 142.0, 145.5, 150.0, 156.5, 163.7.

Anal. Calcd for $C_{23}H_{21}NO_4S$: C, 67.79; H, 5.19; N, 3.44%. Found: C, 67.9; H, 5.3; N, 3.4%.

(*E*) 3-(4-Methylphenyl)-5-(2-thienyl)-2-cyclohexen-1-one Oxime Tosylate (**3j**).

This compound was obtained as colorless crystals (alcohol), yield 92%, mp 92-93 ºC; ir (KBr) 3039, 2930, 2860, 1615, 1499, 1378, 1180, 1168, 1090 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.36 (s, 3 H), 2.44 (s, 3 H), 2.57 (dd, *J* = 18.0, 12.9 Hz, 1 H), 2.75 (ddd, *J* = 17.4, 9.9, 2.1 Hz, 1 H), 3.03 (dd, *J* = 17.4, 4.2 Hz, 1 H), 3.36-3.44 (m, 2 H), 6.56 (d, *J* = 2.1 Hz, 1 H), 6.89 (d, *J* = 3.0 Hz, 1 H), 6.95 (dd, *J* = 4.8, 5.1 Hz, 1 H), 7.15-7.37 (m, 7 H), 7.89 (d, *J* = 8.1 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 21.6, 22.1, 31.7, 34.8, 36.6, 117.7, 124.0, 125.9, 127.3, 129.3, 129.8, 129.9, 130.0, 133.1, 136.3, 139.9, 145.4, 147.4, 150.2, 163.9.

Anal. Calcd for $C_{24}H_{23}NO_3S_2$: C, 65.88; H, 5.30; N, 3.20%. Found: C, 65.8; H, 5.4; N, 3.3%.

Preparation of 6-amino-3,5-diaryl-2-cyclohexen-1-one Hydrochloride (**4**).

General Procedure.

A suspension of 3,5-diaryl cyclohexen-1-one oxime tosylate **3** (8 mmol) in absolute alcohol (50 mL) was stirred with 0.3 g (13 mmol) of sodium in absolute alcohol (20 mL) for 8 h. After completion of the reaction, the mixture was diluted with water, acidified with acetic acid (2 mL) and extracted with ether. The ethereal solution was washed with water. The amine hydrochloride was isolated by extracting the ether solution several times with 10% hydrochloric acid followed by evaporating the acid solution under reduced pressure and the solid obtained was washed with alcohol. All the amine hydrochlorides have their melting points above 250 ºC.

Preparation of N-(2-oxo-4,6-diaryl-3-cyclohexenyl)acetamide (**5**).

General Procedure.

To a solution of amine hydrochloride (**4**) taken as such from the previous stage in water (50 mL) was added sodium acetate (3 g) in water (10 mL) and warmed on a water bath at 70 ºC. To this suspension about 3 mL of acetic anhydride was added with constant stirring and the stirring was continued for further 30 min. The separated acetate was filtered, washed with water and dried. The unreacted amine hydrochloride was removed by dissolving the acetate in chloroform and filtering off the suspended hydrochloride. The filtrate was evaporated and recrystallised from alcohol to get **5**.

N-(2-Oxo-4,6-diphenyl-3-cyclohexenyl)acetamide (**5a**).

This compound was obtained as pale yellow crystals (alcohol), yield 61%, mp 149-150 ºC; ir (KBr) 3276, 3058, 2927, 1671, 1646, 1540, 1423, 1371, 1243, 1141 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 1.86 (s, 3 H), 3.09-3.23 (m, 2 H), 3.32-3.46 (m, 1 H), 5.12 (dd, *J* = 13.2, 9.0 Hz, 1 H), 5.74 (d, *J* = 8.1 Hz, 1 H), 6.57 (s, 1 H), 7.26-7.54 (m, 10 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 24.2, 39.0, 48.8, 59.4, 124.9, 127.3, 128.7, 128.9, 129.9, 130.0, 131.6, 138.6, 141.3, 159.3, 171.8, 197.5.

Anal. Calcd for $C_{20}H_{19}NO_2$: C, 78.66; H, 6.27; N, 4.59%. Found: C, 78.6; H, 6.3; N, 4.7%.

N-[4-(4-Chlorophenyl)-2-oxo-6-phenyl-3-cyclohexenyl]-acetamide (**5b**).

This compound was obtained as pale yellow crystals (alcohol), yield 60%, mp 88-89 ºC; ir (KBr) 3280, 3051, 2925, 1669, 1645, 1535, 1418, 1368, 1240, 1140 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 1.86 (s, 3 H), 3.03-3.07 (m, 2 H), 3.36-3.42 (m, 1 H), 5.04 (dd, *J* = 13.5, 8.7 Hz, 1 H), 5.85 (d, *J* = 8.7 Hz, 1 H), 6.49 (d, *J* = 1.5 Hz, 1 H), 7.27-7.59 (m, 9 H). ^{13}C NMR (75 MHz, $CDCl_3$) δ 23.0, 37.6, 47.3, 58.2, 123.9, 127.4, 127.7, 128.7, 128.8, 129.1, 135.8, 136.6, 139.9, 156.6, 170.7, 196.0.

Anal. Calcd for $C_{20}H_{18}ClNO_2$: C, 70.69; H, 5.34; N, 4.12%. Found: C, 70.8; H, 5.4; N, 4.2%.

N-[4-(4-Chlorophenyl)-6-(4-methoxyphenyl)-2-oxo-3-cyclohexenyl]acetamide (**5c**).

This compound was obtained as pale yellow crystals (alcohol), yield 65%, mp 90-91 ºC; ir (KBr) 3295, 3058, 2918, 1675, 1670, 1530, 1423, 1375, 1245, 1144 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 1.88 (s, 3 H), 2.97-3.13 (m, 2 H), 3.29-3.38 (m, 1 H), 3.79 (s, 3 H), 5.03 (dd, *J* = 12.6, 8.7 Hz, 1 H), 5.71 (d. *J* = 8.7 Hz, 1 H), 6.52 (s, 1 H), 6.87 (d, *J* = 8.1 Hz, 2 H), 7.22-7.55 (m, 6 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 23.4, 38.4, 47.2, 55.7, 58.9, 114.5, 124.3, 127.9, 129.1, 129.5, 132.4, 136.3, 137.0, 157.1, 159.3, 171.1, 196.6.

Anal. Calcd for $C_{21}H_{20}ClNO_3$: C, 68.20; H, 5.45; N, 3.79%. Found: C, 68.1; H, 5.4; N, 3.9%.

N-[4-(2-Naphthyl)-2-oxo-6-phenyl-3-cyclohexenyl]acetamide (**5d**).

This compound was obtained as pale yellow crystals (alcohol), yield 62%, mp 90-91ºC; ir (KBr) 3298, 3040, 2928, 1664, 1660, 1535, 1420, 1380, 1251, 1149 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 1.88 (s, 3 H), 3.18-3.28 (m, 2 H), 3.37-3.49 (m, 1 H), 5.16 (dd, *J* = 13.2, 9.0 Hz, 1 H), 5.71 (d, *J* = 9.0 Hz, 1 H), 6.72 (s, 1 H), 7.11-7.91 (m, 12 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 23.1, 37.8, 47.7, 58.3, 123.1, 123.9, 126.5, 126.9, 127.5, 127.6, 127.7, 127.8, 128.6, 128.7, 128.8, 133.0, 134.1, 134.5, 140.2, 157.6, 170.6, 196.3.

Anal. Calcd for $C_{24}H_{21}NO_2$: C, 81.10; H, 5.96; N, 3.94%. Found: C, 81.2; H, 6.0; N, 3.9%.

N-[6-(4-Methylphenyl)-2-oxo-4-phenyl-3-cyclohexenyl]-acetamide (**5e**).

This compound was obtained as pale yellow crystals (alcohol), yield 61%, mp 97-98 ºC; ir (KBr) 3295, 3056, 2921, 1654, 1610, 1545, 1446, 1371, 1250, 1141 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 1.86 (s, 3 H), 2.33 (s, 3 H), 3.00-3.17 (m, 2 H), 3.30-3.41 (m, 1 H), 5.08 (dd, *J* = 11.7, 8.1 Hz, 1 H), 5.85 (d, *J* = 8.1 Hz, 1 H), 6.54 (s, 1 H), 7.00-7.58 (m, 9 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 22.7, 24.6, 39.6, 48.8, 59.9, 125.3, 127.8, 129.2, 130.5, 131.0, 132.1, 138.7, 138.8, 139.1, 159.9, 172.4, 198.1.

Anal. Calcd for $C_{21}H_{21}NO_2$: C, 78.97; H, 6.63; N, 4.39%. Found: C, 78.9; H, 6.6; N, 4.5%.

N-[6-(4-Methoxyphenyl)-2-oxo-4-phenyl-3-cyclohexenyl]-acetamide (**5f**).

This compound was obtained as pale yellow crystals (alcohol), yield, 65%, mp 95-96 ºC; ir (KBr) 3282, 3048, 2919, 1663, 1640, 1530, 1420, 1362, 1235, 1142 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 1.86 (s, 3 H) 3.07-3.16 (m, 2 H) 3.28-3.38 (m, 1 H), 3.78 (s, 3 H), 5.05 (dd, *J* = 13.2, 8.7 Hz, 1 H), 5.92 (d, *J* = 8.7 Hz, 1 H), 6.53 (s, 1 H), 6.86 (d, *J* = 8.4 Hz, 2 H), 7.22-7.57 (m, 7 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 23.4, 38.4, 47.3, 55.6, 58.9, 114.4, 124.1, 126.6, 129.2, 129.3, 130.8, 132.7, 138.0, 158.7, 159.2, 171.1, 196.9.

Anal. Calcd for $C_{21}H_{21}NO_3$: C, 75.20; H, 6.31; N, 4.18%. Found: C, 75.3; H, 6.4; N, 4.3%.

N-[6-(2-Furyl)-2-oxo-4-(2-thienyl)-3-cyclohexenyl]acetamide (**5g**) with its Fully Aromatized Compound.

This mixture was obtained as viscous liquid, yield 59%.

N-[6-(4-Methylphenyl)-2-oxo-4-(2-thienyl)-3-cyclohexenyl]-acetamide (**5h**).

This compound was obtained as pale yellow crystals (alcohol), yield 60%, mp 76-77 ºC; ir (KBr) 3280, 3052, 2910, 1661, 1642, 1533, 1424, 1366, 1238, 1143 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 1.85 (s, 3 H), 2.34 (s, 3 H), 3.02-3.22 (m, 2 H), 3.32-3.41 (m, 1 H), 5.04 (dd, *J* = 12.9, 8.7 Hz, 1 H), 5.82 (d, *J* = 8.7 Hz, 1 H), 6.53 (s, 1 H), 6.99-7.51 (m, 7 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 21.5, 22.4, 38.0, 47.2, 58.7, 121.5, 128.0, 128.5, 128.9, 129.8, 130.0, 137.4, 137.6, 142.0, 151.5, 171.1, 196.3.

Anal. Calcd for $C_{19}H_{19}NO_2S$: C, 70.12; H, 5.88; N, 4.30%. Found: C, 70.0; H, 6.0; N, 4.2%.

N-[6-(2-furyl)-2-oxo-4-phenyl-3-cyclohexenyl]acetamide (**5i**) with its Fully Aromatized Compound.

This mixture was obtained as viscous liquid, yield: 58%.

N-[4-(4-Methylphenyl)-2-oxo-6-(2-thienyl)-3-cyclohexenyl]-acetamide (**5j**).

This compound was obtained as pale yellow crystals (alcohol), yield 62%, mp 105-106 ºC; ir (KBr) 3300, 3056, 2925, 1662, 1658, 1525, 1420, 1365, 1243, 1142 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.05 (s, 3 H), 2.39 (s, 3 H), 3.16-3.28 (m, 2 H), 3.67-3.77 (m, 1 H), 4.79 (dd, *J* = 12.9, 8.7 Hz, 1 H), 6.13 (d, *J* = 8.7 Hz, 1 H), 6.55 (d, *J* = 2.7 Hz, 1 H), 6.76 (d, *J* = 3.3 Hz, 1 H), 6.84 (dd, *J* = 3.6, 3.3 Hz, 1 H), 7.18-7.49 (m, 5 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 21.8, 23.7, 35.3, 40.4, 59.3, 123.8, 125.0, 126.7, 126.8, 126.9, 130.1, 135.3, 141.2, 141.6, 158.1, 170.9, 194.8.

Anal. Calcd for $C_{19}H_{19}NO_2S$: C, 70.12; H, 5.88; N, 4.30%. Found: C, 70.1; H, 5.9; N, 4.4%.

Reaction of *N*-(2-Oxo-4,6-diaryl-3-cyclohexenyl)acetamide (**5**) with $POCl_3$.

General Procedure.

N-(2-Oxo-4,6-diaryl-3-cyclohexenyl)acetamide (**5**) (0.5 g) was refluxed with 3 mL of $POCl_3$ for about 3 h. After completion of the reaction, the excess $POCl_3$ was carefully destroyed with water and the product was extracted with chloroform. The chloroform layer was washed with water, dried and evaporated. The ratios of **6**, **7** and **8** were calculated from the ^1H NMR spectra of the crude mixture (Table 1). The selected ^1H and ^{13}C NMR data of **6** and **7** are given in Tables 2 and 3 respectively. The 4,6-diaryl-2-methyl-1,3-benzoxazole **8** was separated from the mixture by silica column using pet ether- ethyl acetate as eluent.

Dehydrogenation of **6** and **7** by DDQ to 4,6-diaryl-2-methyl-1,3-benzoxazole (**8**).

General Procedure.

To a solution of 1 mmol of **6/7** in anhydrous dioxane (1.5 mL) was added 0.227 g (1 mmol) of 2,3-dichloro-5,6-dicyano-1,4-benzoquinone dissolved in 1.5 mL of anhydrous dioxane. The mixture was heated in an oil bath at 105 ºC for time periods specified below. The mixture was cooled, diluted with water and extracted with ether. The ether solution was washed well with water, dried, evaporated and recrystallised from alcohol to yield pure **8**.

2-Methyl-4,6-diphenyl-1,3-benzoxazole (**8a**).

This compound was obtained as pale yellow crystals (alcohol), reaction time 8 h, yield 85%, mp 141-142 ºC; ir (KBr) 3056, 2919, 2854, 1610, 1579, 1382, 1257, 1116 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.68 (s, 3 H), 7.35-7.53 (m, 6 H), 7.64-7.69 (m, 4 H), 7.95 (dd, *J* = 8.4, 1.2 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 15.1, 108.1, 123.4, 127.9*, 128.3, 129.1, 129.2, 129.3, 133.1, 137.8, 138.8, 139.1, 141.4, 152.5, 164.6 (*two carbon signals are merged together).

Anal. Calcd for $C_{20}H_{15}NO$: C, 84.19; H, 5.30; N, 4.91%. Found: C, 84.3; H, 5.2; N, 4.8%.

6-(4-Chlorophenyl)-2-methyl-4-phenyl-1,3-benzoxazole (**8b**).

This compound was obtained as pale yellow crystals (alcohol), reaction time 9 h, yield 82%, mp 119-120 ºC; ir (KBr) 3048, 2915, 2855, 1614, 1467, 1386, 1249, 1093 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.69 (s, 3 H), 7.41-7.66 (m, 9 H), 7.93 (dd, *J* = 8.4, 1.2 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 15.1, 107.9, 123.2, 128.4, 129.1, 129.1, 129.2, 129.5, 133.2, 134.0, 137.5, 137.6, 139.3, 139.8, 152.5, 64.8.

Anal. Calcd for $C_{20}H_{14}ClNO$: C, 75.12; H, 4.41; N, 4.38%. Found: C, 75.2; H, 4.4; N, 4.5%.

6-(4-Chlorophenyl)-4-(4-methoxyphenyl)-2-methyl-1,3-benzoxazole (**8c**).

This compound was obtained as pale yellow crystals (alcohol), reaction time 8 h, yield 79%, mp 135-136 ºC; ir (KBr) 3050, 2925, 2854, 1610, 1519, 1467, 1376, 1243, 1186, 1033 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.68 (s, 3 H), 3.87 (s, 3 H), 7.05 (d, *J* = 8.7 Hz, 2 H), 7.43 (d, *J* = 8.7 Hz, 2 H), 7.55-7.60 (m, 4 H), 7.90 (d, *J* = 8.7 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 15.1, 55.8, 107.4, 114.6, 122.6, 129.1, 129.4, 130.1, 130.4, 132.9, 133.9, 137.5, 139.1, 139.9, 152.5, 159.9, 164.6.

Anal. Calcd for $C_{21}H_{16}ClNO_2$: C, 72.10; H, 4.61; N, 4.00%. Found: C, 72.2; H, 4.7; N, 3.9%.

2-Methyl-6-(2-naphthyl)-4-phenyl-1,3-benzoxazole (**8d**).

This compound was obtained as pale yellow crystals (alcohol), reaction time 7 h, yield: 87%, mp 160-161 ºC; ir (KBr) 3048, 2915, 2850, 1606, 1577, 1384, 1257, 1114 cm^{-1}. ^1H NMR (300 MHz, $CDCl_3$) δ 2.68 (s, 3 H), 7.37-7.55 (m, 4 H), 7.74-7.99 (m, 9 H), 8.09 (s, 1 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 15.1, 108.3, 123.6, 126.2, 126.5, 126.6, 126.9, 128.1, 128.3, 128.6, 129.0, 129.1, 129.3, 133.1, 133.2, 134.1, 137.8, 138.6, 138.7, 139.2, 152.6, 164.7.

Anal. Calcd for $C_{24}H_{17}NO$: C, 85.94; H, 5.11; N, 4.18%. Found: C, 85.8; H, 5.2; N, 4.3.

2-Methyl-4-(4-methylphenyl)-6-phenyl-1,3-benzoxazole (**8e**).

This compound was obtained as pale yellow crystals (alcohol), reaction time 9 h, yield 86%, mp 131-132 ºC; ir (KBr) 3044, 2923, 2845, 1604, 1575, 1469, 1378, 1253, 1112 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.41 (s, 3 H), 2.67 (s, 3 H), 7.32 (d, *J* = 8.1 Hz, 2 H), 7.36-7.49 (m, 3 H), 7.61-7.68 (m, 4 H), 7.84 (d, *J* = 8.1 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 15.1, 21.7, 107.8, 123.1, 127.8, 127.9, 129.1, 129.3, 129.8, 133.1, 134.9, 138.1, 138.8, 139.0, 141.5, 152.5, 164.5.

Anal. Calcd for $C_{21}H_{17}NO$: C, 84.25; H, 5.72; N, 4.68%. Found: C, 84.1; H, 5.6; N, 4.8%.

4-(4-Methoxyphenyl)-2-methyl-6-phenyl-1,3-benzoxazole (**8f**).

This compound was obtained as pale yellow crystals (alcohol), reaction time 10 h, yield 84%, mp 85-86 ºC; ir (KBr) 3049, 2933, 2848, 1606, 1519, 1469, 1382, 1253, 1182, 1035 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.66 (s, 3 H), 3.85 (s, 3 H), 7.04 (d, *J* = 8.7 Hz, 2 H), 7.36 (td, *J* = 7.5, 2.1 Hz, 1 H), 7.46 (dd, *J* = 7.8, 7.5 Hz, 2 H), 7.58 (d, *J* = 1.8 Hz, 1 H), 7.63-7.66 (m, 3 H), 7.91 (d, *J* = 8.7 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 15.1, 55.8, 107.5, 114.6, 122.8, 127.8, 127.9, 129.3, 130.3, 130.4, 132.7, 138.8, 138.9, 141.5, 152.5, 159.9, 164.4.

Anal. Calcd for $C_{21}H_{17}NO_2$: C, 79.98; H, 5.43; N, 4.44%. Found: C, 80.1; H, 5.2; N, 4.5%.

4-(2-Furyl)-2-methyl-6-(2-thienyl)-1,3-benzoxazole (**8g**).**

This compound was obtained as pale yellow crystals (alcohol), mp 79-80 ºC; ir (KBr) 3048, 2928, 2840, 1610, 1575, 1473, 1375, 1228, 1110, 1008 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.67 (s, 3 H), 6.57 (dd, *J* = 3.6, 1.8 Hz, 1 H), 7.09 (dd, *J* = 5.1, 3.6 Hz, 1 H), 7.30 (dd, *J* = 5.1, 1.2 Hz, 1 H), 7.37 (dd, *J* = 3.6, 1.2 Hz, 1 H), 7.41 (dd, *J* = 3.3, 0.6 Hz, 1 H), 7.55-7.57 (m, 2 H), 7.95 (d, *J* = 1.5 Hz, 1 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 15.1, 106.4, 111.1, 112.5, 117.9, 122.7, 124.0, 125.5, 128.5, 131.9, 136.8, 142.8, 144.4, 150.5, 152.4, 164.7 (** obtained directly from **5**).

Anal. Calcd for $C_{16}H_{11}NO_2S$: C, 68.31; H, 3.94; N, 4.98%. Found: C, 68.2; H, 3.8; N, 4.8%.

2-Methyl-4-(4-methylphenyl)-6-(2-thienyl)-1,3-benzoxazole (**8h**).

This compound was obtained as pale yellow crystals (alcohol), reaction time 7 h, yield 80%, mp 84-85 ºC; ir (KBr) 3051, 2923, 2835, 1604, 1515, 1421, 1384, 1249, 1228, 1110, 1025 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.42 (s, 3 H), 2.67 (s, 3 H), 7.11 (dd, *J* = 5.1, 3.6 Hz, 1 H), 7.30-7.34 (m, 3 H), 7.36 (dd, *J* = 3.6, 1.2 Hz, 1 H), 7.65 (d, *J* = 1.5 Hz, 1 H), 7.70 (d, *J* = 1.5 Hz, 1 H), 7.82 (d, *J* = 8.1 Hz, 2 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 15.1, 21.7, 106.6, 122.1, 123.9, 125.4, 128.5, 129.1, 129.8, 131.8, 133.3, 134.5, 138.3, 139.2, 144.6, 152.4, 164.6.

Anal. Calcd for $C_{19}H_{15}NOS$: C, 74.72; H, 4.95; N, 4.59%. Found: C, 74.8; H, 5.0; N, 4.7%.

4-(2-Furyl)-2-methyl-6-phenyl-1,3-benzoxazole (**8i**).**

This compound was obtained as pale yellow crystals (alcohol), mp 76-77 ºC; ir (KBr) 3047, 2920, 2850, 1666, 1593, 1492, 1359, 1126 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.71 (s, 3 H), 6.59 (dd, *J* = 3.6, 1.8 Hz, 1 H), 7.36-7.56 (m, 6 H), 7.68 (dd, *J* = 7.2, 1.5 Hz, 2 H), 7.95 (d, *J* = 1.5 Hz, 1 H) (** obtained directly from **5**).

Anal. Calcd for $C_{18}H_{13}NO_2$: C, 78.53; H, 4.76; N, 5.09%. Found: C, 78.4; H, 4.8; N, 5.0%.

2-Methyl-6-(4-methylphenyl)-4-(2-thienyl)-1,3-benzoxazole (**8j**).

This compound was obtained as pale yellow crystals (alcohol), reaction time 8 h, yield 86%, mp 175-176 ºC; ir (KBr) 3045, 2921, 2840, 1606, 1571, 1475, 1434, 1396, 1261, 1112, 1043 cm^{-1}; ^1H nmr (300 MHz, $CDCl_3$) δ 2.42 (s, 3 H), 2.71 (s, 3 H), 7.17 (dd, *J* = 5.1, 3.6 Hz, 1 H), 7.29 (d, *J* = 7.8 Hz, 2 H), 7.38 (dd, *J* = 5.1, 0.9 Hz, 1 H), 7.53-7.56 (m, 3 H), 7.76 (d, *J* = 1.2 Hz, 1 H), 7.98 (dd, *J* = 3.6, 0.9 Hz, 1 H); ^{13}C nmr (75 MHz, $CDCl_3$) δ 15.2, 21.5, 107.7, 121.4, 126.2, 127.3, 127.7, 128.4, 130.0, 137.6, 137.8, 137.9, 138.3, 138.9, 140.0, 152.6, 164.5.

Anal. Calcd for $C_{19}H_{15}NOS$: C, 74.72; H, 4.95; N, 4.59%. Found: C, 74.6; H, 5.1; N, 4.4%.

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