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# 3/4-Phenylene Bisheterocycles from Ring Transformation Reaction of Sydnone Derivatives: Synthesis of 3-[3/4-Heterocyclyl]phenyl-5-methyl-3*H*-[1,3,4]-oxadiazol-2-ones from 3/4-Acetylphenylsydnones and Their Biological Properties

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ABSTRACT: *The bifunctional 3/4-[acetyl]phenylsydnones* **1a, 1b** *were subjected to a one-pot ring conversion to 3-[3/4-acetyl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-ones* **2a, 2b**, *which on further bromination yielded the 3-[3/4-bromoacyl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-ones* **3a, 3b**. *Reaction of these compounds with thiourea yielded the 3-[3/4-(2-aminothiazol-4-yl)]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-ones* **4a, 4b**. *The other thiazole derivatives* **5a, 5b–7a, 7b** *were prepared by using thiosemicarbazide, thioacetamide, and thiobenzamide, respectively. In another reaction of the bromoacetyl compounds (***3a, 3b***) with 2-aminopyridine and 2-aminothiazole, the fused biheterocyclic compounds 3-[3/4-imidazo-[1,2-a]pyridine-2-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-ones* **8a, 8b** *and 3-[3/4-imidazo-[2,1-b]-thiazol-6-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-ones* **9a, 9b** *were obtained. The 3-[3/4-(benzofuran-2-carbonyl)]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-ones* **10a, 10b** *were obtained by treatment of compounds* **3a, 3b** *with o-hydroxy benzaldehyde. Most of these compounds exhibited antifungal activity greater than the reference drugs used.* © 2007 Wiley Periodicals, Inc. Heteroatom Chem 18:50–54, 2007; Published online in Wiley InterScience (www.interscience.wiley.com). DOI 10.1002/hc.20255

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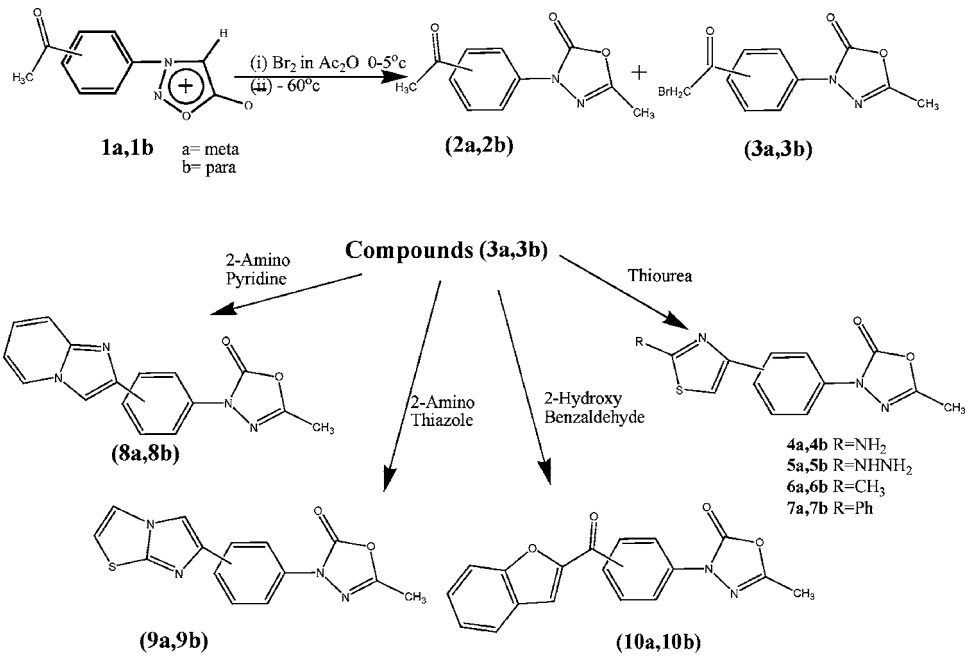
## *INTRODUCTION*

Sydnones readily undergo one-step ring conversion to a variety of heterocycles by 1,3-dipolar cycloaddition reactions [1], and a few such reactions utilizing this latent functionality of sydnones have been reported from our laboratory [2–5]. Sydnones containing appropriate functional groups can act as bifunctional systems, serving as important precursors for the synthesis of bisheterocycles. The synthetic method of bisheterocycles, which is lengthy and affected sequentially, requires bifunctional precursors, which in turn are not readily accessible. In an attempt to exploit the synthetic utility of some bifunctional sydnones and to develop simple, concise, and convenient method for the bisheterocycles, we now report the synthesis of the title compounds from 3/4-[acetyl]phenylsydnones **1a, 1b** using the one-pot 1,3-dipolar cycloaddition reaction. We have

earlier reported [5] the facile one-pot synthesis of 3-aryl-5-methyl-3*H*-[1,3,4]-oxadiazol-2-ones from 3-arylsydnones, and this was the first documentation of their spectral and biological studies. In continuation of this work and to carry out structural modifications, we thought of combining some heterocycles with these oxadiazolinones. Initially, we have selected the thiazole, imidazopyrimidine, imidazothiazole, and the benzofuran heterocycles, not only because of their pronounced biological activities[6–9] but also because of their ease of synthesis from the bromoacetyl moiety. Bisheterocycles, which are mainly of synthetic origin, have received great deal of attention in recent years, as they can form useful molecular models to compare the reactivities of the heterocyclic rings and to evaluate their biological properties.

## *RESULTS AND DISCUSSION*

In the present work, 3/4-[acetyl]phenylsydnones **1a, 1b** were first subjected to ring conversion to 3-[3/4-acetyl]phenyl-5-methyl-3*H*-[1,3,4]-oxadiazol-2-ones **2a, 2b** by one-pot reaction with bromine in acetic anhydride. The formation of these acetyl compounds was accompanied by bromination of the acetyl group to give the 3-[3/4-bromoacyl]phenyl-5-methyl-3*H*-[1,3,4]-oxadiazol-2-ones **3a, 3b**, and the product obtained was a mixture of the acetyl and the bromoacetyl compounds, which could not be separated. The mixture of the acetyl and bromoacetyl derivatives was apparent in the $^1$H NMR spectrum by the presence of $CH_3$ protons of the acetyl group at $\delta$ 2.60 and also the $CH_2$ protons of the bromoacetyl compound at $\delta$ 4.62. This mixture on further bromination using bromine in acetic acid converted the acetyl compounds **2a, 2b**, also to the bromoacetyl derivatives **3a, 3b**. These compounds serve as key intermediates for the preparation of various bisheterocycles **5a, 5b–10a, 10b**. Compounds **3a, 3b** on reaction with thiourea yielded 3-[3/4-(2-aminothiazol-4-yl)]phenyl-5-methyl-3*H*-[1,3,4]-oxadiazol-2-ones **5a, 5b**, which are bisheterocycles containing a biologically potent thiazole ring. Similarly, other thiazole derivatives **6a, 6b–8a, 8b** were prepared by using thiosemicarbazide, thioacetamide, and thiobenzamide, respectively. In another reaction of the bromoacetyl compounds **3a, 3b** with 2-aminopyridine and 2-aminothiazole, the fused bisheterocyclic compounds 3-[3/4-imidazo-[1,2-*a*]pyridine-2-yl]-**9a, 9b** and -[3/4-imidazo-[2,1-*b*]thiazol-6-yl]phenyl-5-methyl-3*H*-[1,3,4]-oxadiazol-2-ones **10a, 10b** were obtained. Another bisheterocyclic system, 3-[3/4-(benzofuran-2-carbonyl)]phenyl-5-methyl-3*H*-[1,3,4]-oxadiazol-2-ones **10a, 10b**, where the two heterocyclic rings are separated by a benzoyl group, was prepared by the reaction of the bromoacetyl compounds **3a, 3b** with *o*-hydroxy benzaldehyde in the presence of anhydrous $K_2CO_3$ (Scheme 1).




**SCHEME 1**

All these compounds were characterized by their spectral and analytical data. All the $^1$H NMR spectra of the 4,4′-disubstituted phenyl compounds **2a–10a** exhibited the AA′BB′ splitting pattern for the phenyl ring protons.

### *Antimicrobial Activity*

All the newly synthesized compounds were evaluated for their antibacterial activity against Gram negative (*Escherichia coli*) and Gram positive (*Bacillus cirroflagellosus*) bacteria and their antifungal activity against *Aspergillus niger* and *Fusarium poa*, with Ciprofloxacin and Griseofulvin as reference drugs, respectively. Only the *p*-bromoacetyl compound **3a** showed growth-inhibitory action two times more than the reference drug against *E. coli*, *A. niger*, and *F. poa*, while the corresponding *m*-isomer (**3b**) was weak to moderately active against all these microbes. The 2-aminothiazole derivative (**5a**) exhibited considerable selective bacterial growth inhibition only against *E. coli* and moderate activity against both the fungi. In contrast, the presence of a 2-hydrazine group (**6a**) reduced the antibacterial activity against both the strains, while the 2-methyl- (**6a**) and the 2-phenyl- (**7a**) substituted compounds showed weak antibacterial activity. All the corresponding *m*-isomers were weakly active against all the microbes. In view of the variation of the biological activity observed for the isomeric compounds, the synthesis of structurally modified systems is continued and is in progress.

## *EXPERIMENTAL*

IR spectra were recorded on a Nicolet-Impact 410 FT-IR spectrophotometer in KBr pellets. $^1$H NMR spectra were recorded on a Brucker-Varian 300 MHz FT-NMR spectrophotometer in $CDCl_3$ using TMS as an internal standard. Purity of the compounds was checked by TLC on silica gel plates.

The synthesis of 3-[3/4-acetyl]phenyl sydnones **1a, 1b** [10] and the sydnone ring transformation of these compounds to the 3-[3/4-acetyl]-/3-[4-bromoacyl]phenyl-5-methyl-3*H*-[1,3,4]-oxadiazol-2-ones **2a, 2b** and **3a, 3b** was carried out by literature methods [11].

### *3-[3/4-Bromoacyl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-ones* **3a, 3b**

The above mixture of compounds **2a**/**2b** and **3a**/**3b** (0.01 mol) was suspended in glacial acetic acid (10 mL), and bromine (0.5 mL, 0.11 mol) in acetic acid (5 mL) was added with stirring at room temperature. After complete addition, stirring was continued for 30 min. The reaction mixture was poured into ice water, the solid separated was filtered, dried, and crystallized from methanol to obtain compounds **3a, 3b**.

*3-[3-Bromoacyl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **3a**. White crystals, m.p. 141–143°C, yield 81%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 7.83–7.60 (m, 4H, Ar-H), 4.60 (s, 2H, $CH_2$), 2.35 (s, 3H, oxadiazolinone $CH_3$); IR 1770 (lactone $\nu_{C=O}$), 1675 (acetyl $\nu_{C=O}$). Calculated for $C_{11}H_9N_2O_3Br$(297): C, 44.46%; H, 3.03%; N, 9.48%. Found: C, 44.08%; H, 2.98%; N, 9.01%.

*3-[4-Bromoacyl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **3b**. White crystals, m.p. 162–64°C, yield 85%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 8.12–8.00 (4H, Ar-H), 4.44 (s, 2H, $CH_2$), 2.39 (s, 3H, oxadiazolinone $CH_3$); IR 1776 (lactone $\nu_{C=O}$), 1671 (acetyl $\nu_{C=O}$). Calculated for $C_{11}H_9N_2O_3Br$(297): C, 44.46%; H, 3.03%; N, 9.48%. Found: C, 44.06%; H, 3.00%; N, 9.00%.

### *3-[3/4-(2-Substituted-thiazol-4-yl)]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-ones* **4a, 4b–7a, 7b**

*General Procedure.* 3-[3/4-Bromoacyl]phenyl-5-methyl-3*H*-[1,3,4]-oxadiazol-2-one **3a**/**3b** (0.01 mol) was dissolved in ethanol (10 mL), and thiourea (0.5 g, 0.01 mol) was added with stirring at room temperature. The mixture was stirred for 30 min and the solution was made alkaline with sodium bicarbonate solution. The solid separated was filtered, dried, and crystallized from ethanol to get compounds **4a**/**4b**. Similarly, compounds **5a**/**5b**, **6a**/**6b**, and **7a**/**7b** were prepared using thiosemicarbazide, thioacetamide, and thiobenzamide, respectively.

*3-[3-(2-Aminothiazol-4-yl)]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **4a**. Yellow crystals, m.p. 151–154°C, yield 81%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 8.01–7.80 (4H, Ar-H), 7.82 (s, 1H, thiazole C5-H), 5.01 (s, 2H, $NH_2$, $D_2O$ exchanged), 2.35 (s, 3H, oxadiazolinone $CH_3$); IR 3370–3350 (br, $\nu_{NH_2}$ of thiazole), 1760 (lactone $\nu_{C=O}$), 1648 ($\nu_{C=N}$). Calculated for $C_{12}H_{10}N_4O_2S$(274): C, 52.55%; H, 3.64%; N, 20.43%. Found: C, 52.08%; H, 3.21%; N, 20.01%.

*3-[4-(2-Aminothiazol-4-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **4b**. Yellow crystals, m.p. 170–172°C, yield 89%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 8.12–8.01 (4H, Ar-H), 7.8 (s, 1H, thiazole C5-H),

4.93 (s, 2H, $NH_2$, $D_2O$ exchanged), 2.33 (s, 3H, oxadiazolinone $CH_3$); IR 3375–3350 ($\nu_{NH_2}$ of thiazole), 1757 (lactone $\nu_{C=O}$), 1652 ($\nu_{C=N}$). Calculated for $C_{12}H_{10}N_3O_2$(260): C, 52.55%; H, 3.64%; N, 20.43%. Found: C, 52.15%; H, 3.20%; N, 20.00%.

*3-[3-(2-Hydrazinothiazol-4-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **5a**. Yellow crystals, m.p. 191–194°C, yield 74%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 8.91 (br, 2H, $NH_2$, $D_2O$ exchanged), 8.15–8.01 (4H, Ar-H), 7.82 (s, 1H, thiazole C5-H), 4.82 (br, 1H, NH, $D_2O$ exchanged), 2.32 (s, 3H, oxadiazolinone $CH_3$); IR 3375–3350 (br, $\nu_{NHNH_2}$), 1751 (lactone $\nu_{C=O}$), 1647 ($\nu_{C=N}$). Calculated for $C_{12}H_{11}N_5O_2S$(289): C, 49.82%; H, 3.80%; N, 24.22%. Found: C, 49.40%; H, 3.31%; N, 23.77%.

*3-[4-(2-Hydrazinothiazol-4-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **5b**. Yellow crystals, m.p. 157–159°C, yield 80%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 8.90 (br, 2H, $NH_2$, $D_2O$ exchanged), 8.15–8.01 (4H, Ar-H), 7.85 (s, 1H, thiazole C5-H), 4.82 (br, 1H, NH, $D_2O$ exchanged), 2.32, 7.85 (s, 1H, thiazole C5-H), 2.32 (s, 3H, oxadiazolinone $CH_3$); IR 3375–3350 (br, $\nu_{NHNH2}$), 1751 (lactone $\nu_{C=O}$), 1647 ($\nu_{C=N}$). Calculated for $C_{12}H_{11}N_5O_2S$(289): C, 49.82%; H, 3.80%; N, 24.22%. Found: C, 49.51%; H, 3.42%; N, 23.97%.

*3-[3-(2-Methylthiazol-4-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **6a**. White crystals, m.p. 191–193°C, yield 71%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 8.10–8.00 (4H, Ar-H), 7.80 (s, 1H, thiazole C5-H), 4.24 (s, 3H, thiazole $CH_3$), 2.33 (s, 3H, oxadiazolinone $CH_3$); IR 1755 (lactone $\nu_{C=O}$), 1648 ($\nu_{C=N}$). Calculated for $C_{13}H_{13}N_3O_2S$(275): C, 56.72%; H, 4.72%; N, 15.27%. Found: C, 56.28%; H, 4.28%; N, 14.724%.

*3-[4-(2-Methylthiazol-4-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **6b**. White crystals, m.p. 180–182°C, yield 87%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 7.95–7.65 (4H, Ar-H), 7.80 (s, 1H, thiazole C5-H), 4.22 (s, 3H, thiazole $CH_3$), 2.33 (s, 3H, oxadiazolinone $CH_3$); IR 1764 (lactone $\nu_{C=O}$), 1648 ($\nu_{C=N}$). Calculated for $C_{13}H_{13}N_3O_2S$(275): C, 56.72%; H, 4.72%; N, 15.27%. Found: C, 56.45%; H, 4.38%; N, 14.84%.

*3-[3-(2-Phenylthiazol4-yl]phenyl-5-methyl-3H-[1, 3,4]-oxadiazol-2-one* **7a**. Colorless crystals, m.p. 151–153°C, yield 67%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 7.90–8.10 (m, 9H, Ar-H), 7.85 (s, 1H, thiazole C5-H), 2.33 (s, 3H, oxadiazolinone $CH_3$); IR 1777 (lactone $\nu_{C=O}$), 1643 ($\nu_{C=N}$). Calculated for $C_{16}H_{15}N_3O_2S$(377): C, 67.90%; H, 3.97%; N, 11.40%. Found: C, 67.68%; H, 3.49%; N, 11.01%.

*3-[4-(2-Phenylthiazol-4-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **7b**. Colorless crystals, m.p. 184–186°C, yield 73%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 8.15–7.60 (m, 9H, Ar-H), 7.85 (s, 1H, thiazole C5-H), 2.39 2.32 (s, 3H, oxadiazolinone $CH_3$); IR 1768 (lactone $\nu_{C=O}$), 1645 ($\nu_{C=N}$). Calculated for $C_{16}H_{15}N_3O_2S$(377): C, 67.90%; H, 3.97%; N, 11.40%. Found: C, 67.50%; H, 3.55%; N, 11.02%.

### *3-[3/4-Imidazo-[1,2-a]pyridine-2-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-ones* **8a, 8b**

A mixture of compound **3a**/**3b** (0.01 mol) and 2-aminopyridine (0.01 mol) in methanol (20 mL) was refluxed on a water bath for 2 h. The reaction was followed by TLC. After the completion of the reaction, the solution was diluted with water and the resulting mixture, containing a yellow solid, was cooled in an ice bath. The solid separated was filtered, washed thoroughly with water, and dried to obtain compounds **8a, 8b**.

*3-[3-Imidazo-[1,2-a]pyridine-2-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **8a**. Yellow crystals, m.p. 191–193°C, yield 61%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 8.15–7.60 (m, 9H, Ar-H), 7.85 (s, 1H, thiazole C5-H), 2.39 2.32 (s, 3H, oxadiazolinone $CH_3$); IR 1777 (lactone $\nu_{C=O}$), 1623 ($\nu_{C=N}$). Calculated for $C_{16}H_{12}N_4O_2$(292): C, 65.75%; H, 4.11%; N, 19.17%. Found: C, 65.71%; H, 4.06%; N, 19.12%.

*3-[4-Imidazo-[1,2-a]pyridine-2-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **8b**. Yellow crystals, m.p. 176–178°C, yield 78%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 8.35 (s, 1H, imidazo CH), 7.90–7.65 (8H, Ar-H), 2.31 (s, 3H, oxadiazolinone $CH_3$); IR 1770 (lactone $\nu_{C=O}$), 1603 ($\nu_{C=N}$). Calculated for $C_{16}H_{12}N_4O_2$(292): C, 65.75%; H, 4.11%; N, 19.17%. Found: C, 65.47%; H, 3.85%; N, 18.71%.

### *3-[3/4-Imidazo-[2,1-b]thiazol-6-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-ones* **9a, 9b**

A mixture of compound **3a**/**3b** (0.01 mol) and 2-aminothiazole (0.01 mol) in methanol (20 mL) was refluxed on a water bath for 2 h. After the completion of the reaction, the solution was diluted with water and the resulting mixture, containing a white solid, was cooled in an ice bath. The solid separated was filtered, washed thoroughly with water and dried to obtain compounds **9a, 9b**.

*3-[3-Imidazo-[2,1-b]thiazol-6-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **9a**. White crystals, m.p. 179–181°C, yield 86%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 8.35 (s, 1H, imidazo CH), 7.91–7.53 (4H, Ar-H), 6.81 (d, 1H, $J = 7.9$ Hz, thiazole C4-H), 6.72 (d, 1H, $J = 7.9$ Hz, thiazole C5-H), 2.41 (s, 3H, oxadiazolinone $CH_3$); IR 1770 (lactone $\nu_{C=O}$), 1647 ($\nu_{C=N}$). Calculated for $C_{14}H_{10}N_4O_2S$(298): C, 56.38%; H, 3.55%; N, 18.79%. Found: C, 56.02%; H, 2.94%; N, 18.16%.

*3-[4-Imidazo-[2,1-b]thiazol-6-yl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **9b**. White crystals, m.p. 192–195°C, yield 81%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 8.50, (s, 1H, imidazo CH), 7.91–7.53 (4H, Ar-H), 6.85 (d, 1H, $J = 8$ Hz, thiazole C4-H), 6.75 (d, 1H, $J = 8$ Hz, thiazole C5-H), 2.41 (s, 3H, oxadiazolinone $CH_3$); IR 1770 (lactone $\nu_{C=O}$), 1647 ($\nu_{C=N}$). Calculated for $C_{14}H_{10}N_4O_2S$(298): C, 56.38%; H, 3.55%; N, 18.79%. Found: C, 56.00%; H, 2.85%; N, 18.46%.

### *3-[3/4-(Benzofuran-3-carbonyl)]]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-ones* **10a, 10b**

Compound **3a**/**3b** (0.05 mol) was dissolved in dry ethanol (50 mL) and cooled to 10–15°C. 2-Hydroxy benzaldehyde (0.05 mol) and anhydrous $K_2CO_3$ (1 g) were added. The reaction mixture was stirred for 1 h at room temperature and then refluxed on a water bath for 30 min. It was then cooled and poured into an ice water and the yellow solid obtained was filtered to obtain compounds **10a, 10b**.

*3-[3-(Benzofuran-2-carbonyl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **10a**. Yellow crystals, m.p. 154–156°C, yield 72%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 8.22–8.15 (4H, Ar-H), 7.77–7.30 (m, 4H, benzofuran Ar-H), 7.25 (s, 1H, benzofuran C2-H), 2.59 (s, 3H, oxadiazolinone $CH_3$); IR 1782 (lactone $\nu_{C=O}$), 1634 ($\nu_{C=O}$ flanked by benzofuran and phenyl ring), 1597 ($\nu_{C=N}$). Calculated for $C_{18}H_{12}N_2O_4$(320): C, 67.51%; H, 3.75%; N, 8.75%. Found: C, 67.04%; H, 3.28%; N, 8.11%.

*3-[4-(Benzofuran-2-carbonyl]phenyl-5-methyl-3H-[1,3,4]-oxadiazol-2-one* **10b**. Yellow crystals, m.p. 167–169°C, yield 72%. $^1$H NMR ($CDCl_3$, 300 MHz): $\delta$ 8.30–8.21 (4H, Ar-H), 7.77–7.30 (m, 4H, benzofuran Ar-H), 7.30 (s, 1H, benzofuran C2-H), 2.59 (s, 3H, oxadiazolinone $CH_3$); IR 1782 (lactone $\nu_{C=O}$), 1634 ($\nu_{C=O}$ flanked by benzofuran and phenyl ring), 1597 ($\nu_{C=N}$). Calculated for $C_{18}H_{12}N_2O_4$(320): C, 67.51%; H, 3.75%; N, 8.75%. Found: C, 67.15%; H, 3.22%; N, 8.33%.

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