Synthesis of Novel Naphth[1,2-f][1,4]oxazepine-3,4-dione Heterocycles

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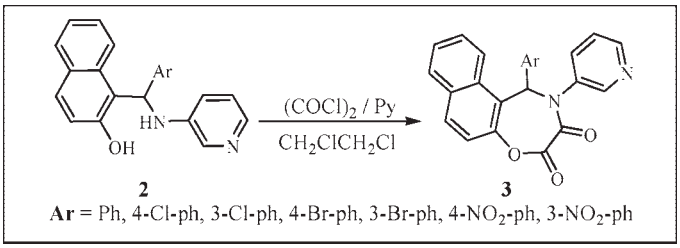

A number of *N*-heteroaryl aminonaphthols (Betti bases) were prepared from the reaction of 2-naphthol, 3-aminopyridine, and aromatic aldehydes. Subsequent condensation of the prepared Betti bases with oxalyl chloride afforded the novel naphth[1,2-f][1,4]oxazepine-3,4-dione heterocycles in moderate to high yields.

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INTRODUCTION

Although Betti's classical procedure for the preparation of 1-(α-aminobenzyl)-2-naphthol (Betti base) was published more than a century ago [1], the possibilities of the application of this versatile synthon in the ring-closure reactions to give naphthalene-condensed heterocyclic derivatives have not been thoroughly investigated. A few publications that have been appeared on this topic focus on the transformation reactions of the Betti base analog aminonaphthols with phosgene, ethyl benzimidate, 2-carboxybenzaldehyde, levulinic acid, salicylaldehyde/acetaldehyde, salicylaldehyde/formalin, benzaldehyde derivatives, and bis-aldehydes/$NaBH_3CN$ to naphthoxazine derivatives [2–6]. Traditionally, the Betti base derivatives synthesis is carried out in organic solvents, such as, EtOH, MeOH, and Et_2O, at room temperature for long time or thermally under solvent-less conditions. As such, utilization of environmental friendly water as solvent not only provides the product in an easy work-up procedure but also is in accord with green sustainable chemistry principles [7–13].

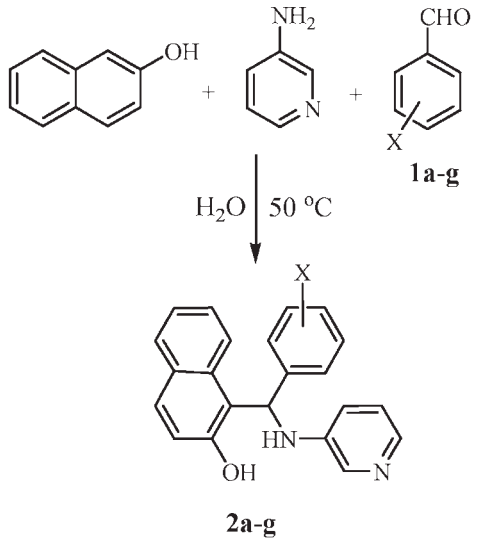
There are many methods for the synthesis of oxazepine ring systems [14]. In the reported synthetic approaches to the aryl-fused derivatives of 1,4-oxazepines, several bifunctional keto acids were used in Ugi condensation reaction [15]. Similarly, a number of 1,4-benzoxazepin-3-ones were obtained via Ugi three-component condensations using bifunctional starting materials containing aldehyde and carboxylic acid [16]. A synthetic procedure was also developed for the preparation of oxazepinedione derivatives by lactonization of some amides obtained from condensation of either 2-acetoxybenzoic acid chloride or the proper acethoxynaphthonic acid chloride with cyclic amino acids [17]. Oxazepine derivatives were described as effective protease inhibitors [18], integrin antagonists, squalene synthase [19], reverse transcriptase inhibitors [20], and antihistamines, which can be used for the efficient therapy of some allergic and dermatological infections [21]. To the best of our knowledge, synthesis of dioxo-1,4-naphthoxazepine via condensation of aminonaphthols (Betti bases) with oxalyl chloride has not been previously reported in literature.

Our very recently reported results on the successful uncatalyzed quantitative preparation of aminonaphthols (Betti bases) via one-pot three-component reaction of 2-naphthol, aromatic aldehydes, and heteroaryl amines, such as 2-aminopyrimidine, 2-aminopyrazine, and 2-aminopyridine [22] prompted us to utilize these bases in the synthesis of the novel dioxo-1,4-naphthoxazepines.

RESULTS AND DISCUSSION

Condensation of Betti bases containing the heteroaryls, such as 2-aminopyrimidine, 2-aminopyrazine, and 2-aminopyridine with oxalyl chloride failed to produce any product with the oxazepinedione structure. It was concluded that the Betti bases containing the very poor electron nature of 2-aminopyrimidine, 2-aminopyrazine, and 2-aminopyridine might have been responsible for

Scheme 1



X = **a**: H, **b**: 4-Cl, **c**: 3-Cl, **d**: 4-Br, **e**: 3-Br, **f**: 4-NO_2, **g**: 3-NO_2

the lack of expected reactivity. Therefore, it was decided to prepare the Betti bases obtained from one-pot condensation of 3-aminopyridine, 2-naphthol with different aromatic aldehydes. These aminonaphthols containing the 3-aminopyridine moiety with partially less electron deficiency with respect to the previously prepared heteroaryl amines were expected to successfully furnish the relevant oxazepine derivatives.

As the model reaction, the one-pot reaction of benzaldehyde, 2-naphthol, and 3-aminopyridine was carried out under our previously reported conditions. It was found that at least 2 h is needed for reaction to be completed at room temperature. The best result was obtained at 50°C, because the reaction was completed during 5 min. Therefore, three-component reactions of 2-naphthol, 3-aminopyridine, and aromatic aldehydes **1a–g** in water afforded aminonaphthol derivatives **2a–g** with excellent yields (Scheme 1). Reaction times and yields of the synthesized Betti base derivatives **2a–g** are presented in Table 1.

Identification of **2a–g** was carried out on the basis of spectroscopic information and elemental analysis. The ^{1}H NMR spectra of compounds **2a–g** show a sharp singlet for the hydroxyl group at δ 10.2 ppm, a doublet for NH proton at δ 6.7 ppm, and a doublet for methine proton at δ 6.5 ppm. D_2O is traditionally added into the NMR tube of compounds containing the OH or NH group to find the relevant signals and their coupling effects. Upon addition of D_2O into the **2a** NMR sample tube, the δ 10.2 ppm and δ 6.7 ppm signals disappeared and the proton of methine located at δ 6.5 ppm moiety collapsed into a singlet. The IR spectra of compounds **2a–g** display two absorption bands at 3373–3406 cm^{-1} for OH and NH groups. Compounds **2a–g** exhibit the expected parent ion peaks with medium intensity in the Mass spectra.

When the newly synthesized *N*-heteroaryl-substituted aminonaphthols **2a–g** were treated with oxalyl chloride in the presence of pyridine in dry 1,2-dichloroethane as solvent, the corresponding novel naphthoxazepine-condensed 1,4-naphthoxazepine-3,4-diones **3a–g** were obtained (Scheme 2).

Identification of **3a–g** was carried out on the basis of ^{1}H NMR, ^{13}C NMR, IR, Mass spectra, and elemental analysis. The absence of OH and NH protons in the ^{1}H NMR and IR spectra confirms the transformation of the starting aminonaphthols into the products by bonding of the oxalyl chloride sp^2 carbons to the hydroxyl and *N*-substituted groups. IR absorption bands appearing at 1633 and 1725 cm^{-1} reveals the presence of lactam and lactone carbonyl groups, respectively. The oxazepine-dione ring methine proton appears as a singlet at δ 6.5 ppm and the other protons are displayed in the aromatic regions. Mass spectra of **3a–g** reveal the presence of the molecular ion peaks and other fragments consistent with the assigned structures.

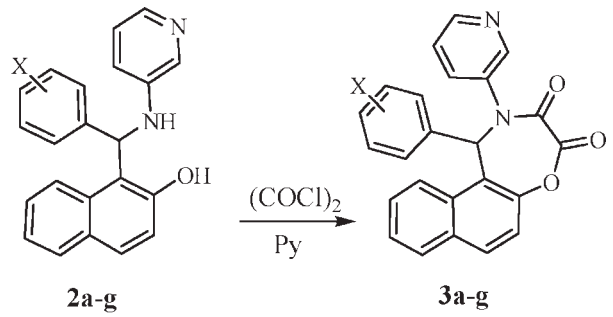
In summary, we have successfully developed a fast, convenient, and efficient method for the synthesis of new *N*-substituted-aminonaphthol (Betti base) derivatives in water. When compared with other previously reported procedures in literatures, this method with the advantages, such as omitting organic solvent, generality

Table 1

Reaction times, yields, and melting points of the products **2** in H_2O.

Compound	Aldehyde	Time (min)	Mp (°C)	Yield (%)
2a	Benzaldehyde	5	187–189	90
2b	4-Chlorobenzaldehyde	25	157–159	92
2c	3-Chlorobenzaldehyde	30	144–145	91
2d	4-Bromobenzaldehyde	50	162–164	95
2e	3-Bromobenzaldehyde	55	149–150	94
2f	4-Nitrrobenzaldehyde	10	179–180	97
2g	3-Nitrrobenzaldehyde	15	190–192	93

Scheme 2



2a-g **3a-g**

X – **a**: H, **b**: 4-Cl, **c**: 3-Cl, **d**: 4-Br, **e**: 3-Br, **f**: 4-NO_2, **g**: 3-NO_2

and simplicity of procedure, lower reaction time, elimination of acid catalyst, and obtaining excellent yields are worth noting. Moreover, we have developed a convenient synthetic strategy to novel naphthoxazepine-condensed derivatives 1,4-naphthoxazepine-3,4-diones. To the best of our knowledge, the synthesis of seven-membered rings consisting of 1,4-naphthoxazepine-3,4-diones have not been previously reported.

EXPERIMENTAL

All commercially available chemicals and reagents were used without further purification. Melting points were determined with an Electrothermal model 9100 apparatus and are uncorrected. IR spectra were recorded on a Shimadzu 4300 spectrophotometer. The ^{1}H and ^{13}C NMR spectra were recorded in DMSO-d_6 on Bruker DRX-500 AVANCE spectrometers. Chemical shifts (δ) are reported in ppm and are referenced to the NMR solvent. Mass spectra of the products were obtained with a HP (Agilent technologies) 5937 Mass Selective Detector. Elemental analyses were carried out by a CHN-O-Rapid Heraeus elemental analyzer (Wellesley, MA).

General procedure for the synthesis of 1-(X-substituted-phenyl(pyridine-3-ylamino)methyl)naphthalene-2-ol (2a–g). To a stirring suspension of 2-naphthol (1.44 g, 10 mmol) in water (15 mL), the appropriate aromatic aldehyde (10 mmol) and 3-aminopyridine (10 mmol) was added. The reaction mixture was stirred at 50°C for the appropriate time (see Table 1). Then water was decanted and the white precipitated product was separated upon addition of ethanol (10 mL) to the mixture with stirring, while cooling to 0–5°C. The precipitate was filtered, washed with cold EtOH, dried, and purified by recrystallization from EtOH to give the colorless crystals of **2a–g**.

1-(Phenyl(pyridine-3-ylamino)methyl)naphthalene-2-ol (2a). IR (potassium bromide): 3394, 3068, 2900, 1625, 1240, 804 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ 6.55 (d, J = 6.6 Hz, 1H, methine-H), 6.72 (d, J = 6.6 Hz, 1H, NH), 7.00–8.15 (m, 15H, NPh-H, Ph-H, and Pyridine-H), 10.25 (s, 1H, OH) ppm; ^{1}H NMR (500 MHz, DMSO-d6 + D_2O): δ 6.57 (s, 1H, methine-H), 7.01–8.09 (m, 15H, NPh-H, Ph-H, and Pyridine-H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 53.39, 119.08, 119.17, 119.56, 123.26, 124.34, 124.94, 126.91, 127.32, 127.59, 129.06, 129.45, 129.70, 130.19, 133.08, 136.73, 138.11, 143.11, 145.44, 153.88 ppm; ms: *m/z* (%) 326 (10) [M$^+$], 231 (100), 202 (50), 94 (40). *Anal.* Calcd for $C_{22}H_{18}N_2O$: C, 80.98; H, 5.52; N, 8.58%. Found: C, 80.87; H, 5.60; N, 8.49%.

1-(4-Chlorophenyl(pyridine-3-ylamino)methyl)naphthalene-2-ol (2b). IR (potassium bromide): 3373, 3051, 2917, 1627, 1299, 806 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ 6.60 (d, J = 6.6 Hz, 1H, methine-H), 6.72 (d, J = 6.6 Hz, 1H, NH), 7.02–8.12 (m, 14H, NPh-H, Ph-H, and Pyridine-H), 10.25 (s, 1H, OH) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 52.86, 119.07, 119.21, 119.26, 123.33, 124.36, 124.69, 127.08, 128.99, 129.42, 129.51, 129.66, 130.39 131.83, 132.95, 136.76, 138.26, 142.27, 145.27, 153.85 ppm; ms: *m/z* (%) 360 (15) [M+], 265 (90), 231 (100), 202 (50), 144 (15), 94 (30). *Anal.* Calcd for $C_{22}H_{17}ClN_2O$: C, 73.23; H, 4.71; N, 7.76%. Found: C, 73.19; H, 4.64; N, 7.80%.

1-(3-Chlorophenyl(pyridine-3-ylamino)methyl)naphthalene-2-ol (2c). IR (potassium bromide): 3404, 3051, 2906, 1625, 1292, 815 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ 6.57 (d, J = 6.8 Hz, 1H, methine-H), 6.74 (d, J = 6.8 Hz, 1H, NH), 7.02–8.13 (m, 14H, NPh-H, Ph-H, and Pyridine-H), 10.28 (s, 1H, OH) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 52.97, 119.10, 119.17, 119.24, 123.39, 124.37, 124.52, 126.29, 127.15, 127.23, 127.32, 129.55, 129.61, 130.48, 130.96, 132.93, 133.81, 136.77, 138.35, 145.16, 145.95, 153.90 ppm; ms: *m/z* (%) 360 (15) [M$^+$], 265 (60), 231 (100), 216 (75), 202 (30), 144 (85), 94 (25). *Anal.* Calcd for $C_{22}H_{17}ClN_2O$: C, 73.23; H, 4.71; N, 7.76%. Found: C, 73.16; H, 4.68; N, 7.72%.

1-(4-Bromophenyl(pyridine-3-ylamino)methyl)naphthalene-2-ol (2d). IR (potassium bromide): 3373, 3051, 2915, 1627, 1249, 806 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ 6.53 (d, J = 6.3 Hz, 1H, methine-H), 6.71 (d, J = 6.3 Hz, 1H, NH), 7.02–8.12 (m, 14H, NPh-H, Ph-H, and Pyridine-H), 10.26 (s, 1H, OH) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 52.93, 119.10, 119.25, 120.33, 123.35, 124.38, 124.60, 124.72, 127.10, 129.52, 129.67, 129.82, 130.41, 131.91, 132.96, 136.78, 138.28, 142.75, 145.29, 153.87 ppm; ms: *m/z* (%) 404 (15) [M$^+$], 406 (15) [M$^+$], 311 (80), 231 (100), 202 (80), 144 (25), 94 (50). *Anal.* Calcd for $C_{22}H_{17}BrN_2O$: C, 65.18; H, 4.19; N, 6.91%. Found: C, 65.21; H, 4.11; N, 6.89%.

1-(3-Bromophenyl(pyridine-3-ylamino)methyl)naphthalene-2-ol (2e). IR (potassium bromide): 3394, 3053, 2923, 1625, 1238, 817 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ 6.59 (d, J = 6.3 Hz, 1H, methine-H), 6.75 (d, J = 6.3 Hz, 1H, NH), 7.02–8.15 (m, 14H, NPh-H, Ph-H, and Pyridine-H), 10.32 (s, 1H, OH) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 52.96, 119.10, 119.15, 119.25, 122.51, 123.39, 124.37, 126.67, 127.21, 129.55, 129.60, 130.13, 130.21, 130.48, 131.25, 131.90, 132.94, 136.78, 138.36, 145.16, 146.19, 153.94 ppm; ms: *m/z* (%) 404 (100) [M$^+$], 406 (100) [M$^+$], 311 (10), 231 (20), 202 (8), 144 (15), 95 (25). *Anal.* Calcd for $C_{22}H_{17}BrN_2O$: C, 65.18; H, 4.19; N, 6.91%. Found: C, 65.14; H, 4.16; N, 6.97%.

1-(4-Nitrophenyl(pyridine-3-ylamino)methyl)naphthalene-2-ol (2f). IR (potassium bromide): 3406, 3087, 2916, 1627, 1514, 1346, 1244, 827 cm^{-1}; ^{1}H NMR (500 MHz, DMSO-d_6): δ 6.68 (d, J = 6.3 Hz, 1H, methine-H), 6.81 (d, J = 6.3 Hz, 1H, NH), 7.06–8.19 (m, 14H, NPh-H, Ph-H, and Pyridine-H), 10.35 (s, 1H, OH) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 53.29, 119.08, 119.11, 119.14, 119.37, 123.47, 124.28, 124.44,

127.37, 128.69, 129.58, 129.64, 130.76, 132.92, 136.86, 138.53, 145.17, 147.01, 151.70, 153.93 ppm; ms: *m/z* (%) 371 (12) [M^+], 260 (30), 231 (100), 202 (50), 144 (15), 94 (22). *Anal*. Calcd for $C_{22}H_{17}N_3O_3$: C, 71.16; H, 4.58; N, 11.32%. Found: C, 71.20; H, 4.57; N, 11.22%.

1-(3-Nitrophenyl(pyridine-3-ylamino)methyl)naphthalene-2-ol (2g). IR (potassium bromide): 3390, 3078, 2925, 1622, 1527, 1348, 1236, 804 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ 6.71 (d, J = 6.7 Hz, 1H, methine-H), 6.93 (d, J = 6.7 Hz, 1H, NH), 7.10–8.23 (m, 14H, NPh-H, Ph-H, and Pyridine-H), 10.40 (s, 1H, OH) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 53.01, 118.75, 119.21, 120.07, 122.09, 122.46, 123.49, 124.22, 124.71, 127.45, 129.59, 129.64, 130.60, 130.80, 132.91, 134.32, 136.07, 137.90, 145.28, 145.81, 148.71, 154.10 ppm; ms: *m/z* (%) 371 (12) [M^+], 276 (50), 260 (65), 231 (100), 202 (75), 144 (90), 94 (85). *Anal*. Calcd for $C_{22}H_{17}N_3O_3$: C, 71.16; H, 4.58; N, 11.32%. Found: C, 71.11; H, 4.50; N, 11.25%.

General procedure for the synthesis of 1-(X-substituted-phenyl)-2-(pyridin-3-yl)-1,2-dihydronaphth[1,2-f][1,4]oxazepine-3,4-diones (3a–g). Oxalyl chloride (1.2 mmol) was added dropwise with ice cooling to a stirring solution of 1-(X-substituted-phenyl(pyridine-3-ylamino)methyl)naphthalene-2-ol **2a–g** (1 mmol) in dry 1,2-dichloroethane (15 mL) containing pyridine (2.4 mmol) under argon atmosphere. The mixture was stirred for 30 min at 0–5°C, then 30 min at room temperature, and refluxed for an additional 45 min. The solvent was then removed under reduce pressure to give a solid compound. It was then triturated in saturated sodium bicarbonate solution (15 mL), filtered, washed with distilled water, and dried. The crude product was recrystallized from EtOH to give colorless crystals of **3a–g**.

1-Phenyl-2-(pyridine-3-yl)-1,2-dihydronaphth[1,2-f][1,4]oxazepine-3,4-dione (3a). Yield: 85%; mp: 232–234°C; IR (potassium bromide): 1718, 1635 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ 6.23 (s, 1H, methine-H), 7.22–8.62 (m, 15H, NPh-H, Ph-H, and Pyridine-H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 65.41, 114.75, 117.35, 122.69, 124.23, 125.86, 128.06, 128.17, 129.18, 129.42, 129.54, 129.73, 131.28, 131.47, 136.41, 137.54, 139.39, 147.53, 149.50, 149.48, 149.53, 150.00 ppm; ms: *m/z* (%) 380 (5) [M^+], 352 (35), 231 (100), 202 (45). *Anal*. Calcd for $C_{24}H_{16}N_2O_3$: C, 75.79; H, 4.21; N, 7.37%. Found: C, 75.70; H, 4.25; N, 7.40%.

1-(4-Chlorophenyl)-2-(pyridine-3-yl)-1,2-dihydronaphth[1,2-f][1,4]oxazepine-3,4-dione (3b). Yield: 83%; mp: 245–246°C; IR (potassium bromide): 1718, 1635 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ 6.73 (s, 1H, methine-H), 7.32–8.55 (m, 14H, NPh-H, Ph-H, and Pyridine-H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 62.81, 115.42, 117.54, 124.06, 124.86, 126.33, 128.44, 129.20, 129.60, 130.03, 130.28, 131.48, 131.66, 131.66, 134.11, 136.61, 137.47, 139.75, 147.31, 149.41, 149.43, 149.94 ppm; ms: *m/z* (%) 415 (10) [M^+], 386 (15), 279 (40), 231 (50), 202 (20), 167 (90), 149 (100). *Anal*. Calcd For $C_{24}H_{15}ClN_2O_3$: C, 69.48; H, 3.62; N, 6.75%. Found: C, 69.51; H, 3.55; N, 6.70%.

1-(3-Chlorophenyl)-2-(pyridine-3-yl)-1,2-dihydronaphth[1,2-f][1,4]oxazepine-3,4-dione (3c). Yield: 76%; mp: 222–223°C; IR (potassium bromide): 1724, 1639 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ 6.74 (s, 1H, methine-H), 7.28–8.56 (m, 14H, NPh-H, Ph-H, and Pyridine-H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 62.82, 115.22, 117.57, 124.03, 124.85, 126.38, 127.00, 128.17, 128.54, 129.20, 129.61, 129.64, 131.49, 131.76, 132.08, 134.33, 136.60, 137.40, 143.09, 147.41, 149.40, 149.43, 149.48, 149.90 ppm; ms: *m/z* (%) 415 (10) [M^+], 386 (50), 265 (90), 231 (100), 202 (50). *Anal*. Calcd For $C_{24}H_{15}ClN_2O_3$: C, 69.48; H, 3.62; N, 6.75%. Found: C, 69.42; H, 3.59; N, 6.80%.

1-(4-Bromophenyl)-2-(pyridine-3-yl)-1,2-dihydronaphth[1,2-f][1,4]oxazepine-3,4-dione (3d). Yield: 80%; mp: 225–227°C; IR (potassium bromide): 1720, 1637 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ 6.71 (s, 1H, methine-H), 7.25-8.54 (m, 14H, NPh-H, Ph-H, and Pyridine-H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 62.88, 115.36, 117.53, 122.77, 124.05, 124.88, 126.33, 128.45, 129.19, 129.60, 130.57, 131.48, 131.67, 132.95, 136.60, 137.47, 140.14, 147.30, 149.40, 149.41, 149.43, 149.92 ppm; ms: *m/z* (%) 458 (5)[M^+], 460 (5) [M^+], 430 (12), 432 (12), 309 (25), 311 (25), 231 (90), 202 (25), 167 (80), 149 (100). *Anal*. Calcd for $C_{24}H_{15}BrN_2O_3$: C, 62.74; H, 3.26; N, 6.10%. Found: C, 62.70; H, 3.33; N, 6.14%.

1-(3-bromophenyl)-2-(pyridine-3-yl)-1,2-dihydronaphth[1,2-f][1,4]oxazepine-3,4-dione (3e). Yield: 78%; mp: 220–222°C; IR (potassium bromide): 1724, 1639 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ 6.73 (s, 1H, methine-H), 7.26–8.56 (m, 14H, NPh-H, Ph-H, and Pyridine-H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 62.78, 115.20, 117.56, 122.92, 124.02, 124.86, 126.39, 127.37, 128.55, 129.18, 129.63, 131.00, 131.49, 131.78, 132.34, 132.52, 136.61, 137.39, 143.28, 147.41, 149.41, 149.45, 149.48, 149.89 ppm; ms: *m/z* (%) 458 (5)[M^+], 460 (5)[M^+], 430 (12), 432 (12), 309 (50), 311 (50), 231 (90), 202 (50), 167 (40), 149 (100). *Anal*. Calcd for $C_{24}H_{15}BrN_2O_3$: C, 62.74; H, 3.26; N, 6.10%. Found: C, 62.80; H, 3.21; N, 6.02%.

1-(4-Nitrophenyl)-2-(pyridine-3-yl)-1,2-dihydronaphth[1,2-f][1,4]oxazepine-3,4-dione (3f). Yield: 78%; mp: 205–207°C; IR (potassium bromide): 1720, 1633, 1519, 1348 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ 6.92 (s, 1H, methine-H), 7.48–8.55 (m, 14H, NPh-H, Ph-H, and Pyridine-H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 62.59, 114.85, 117.61, 123.97, 124.93, 125.25, 126.43, 128.60, 129.15, 129.66, 129.83, 129.85, 131.50, 131.98, 136.64, 137.34, 147.50, 147.68, 148.30, 149.30, 149.57, 149.92 ppm; ms: *m/z* (%) 425 (6) [M^+], 397 (18), 279 (50), 260 (47), 231 (80), 202 (30), 167 (90), 149 (100). *Anal*. Calcd For $C_{24}H_{15}N_3O_5$: C, 67.76; H, 3.53; N, 9.88%. Found: C, 67.80; H, 3.52; N, 9.87%.

1-(3-Nitrophenyl)-2-(pyridine-3-yl)-1,2-dihydronaphth[1,2-f][1,4]oxazepine-3,4-dione (3g). Yield: 80%; mp: 202–203°C; IR (potassium bromide): 1725, 1638, 1524, 1349 cm^{-1}; ^1H NMR (500 MHz, DMSO-d_6): δ 6.95 (s, 1H, methine-H), 7.46–8. (m, 14H, NPh-H, Ph-H, and Pyridine-H) ppm; ^{13}C NMR (125 MHz, DMSO-d_6): δ 62.49, 114.83, 117.60, 123.04, 123.99, 124.57, 124.88, 126.45, 128.65, 129.14, 129.20, 129.67, 131.50, 131.73, 132.02, 134.94, 136.84, 137.23, 142.63, 147.57, 148.91, 149.29, 149.59, 150.09 ppm; ms: *m/z* (%) 425 (5) [M^+], 397 (15), 279 (60), 260 (50), 231 (85), 202 (30), 167 (92), 149 (100). *Anal*. Calcd for $C_{24}H_{15}N_3O_5$: C, 67.76; H, 3.53; N, 9.88%. Found: C, 67.69; H, 3.50; N, 9.92%.

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