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Base-Promoted [4 + 1]/[3 + 1 + 1] Bicyclization for Accessing Functionalized Indeno[1,2-*c*]furans

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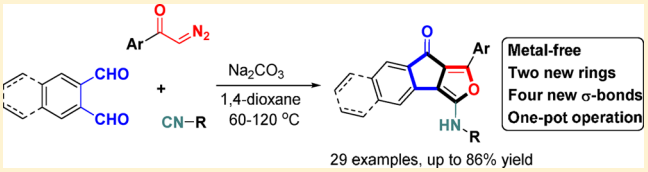
Supporting Information

ABSTRACT: A novel three-component bicyclization strategy for the metal-free synthesis of densely functionalized indeno-[1,2-*c*]furans with generally good yields has been established from readily accessible *o*-phthalaldehydes (OPA), isocyanides, and α-diazoketones. The reaction pathway involves aldol-type addition, 1,2-hydride shift, 5-*exo*-*trig* cyclization, and 1,4-addition as well as an oxo-5-*exo*-*dig* cyclization sequence, resulting in continuous multiple bond-forming events including C−C and C−O bonds to rapidly build up functional oxo-heterocycles.

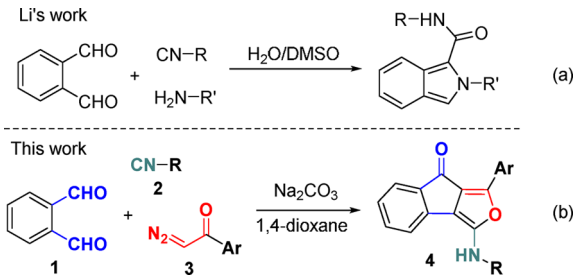


INTRODUCTION

Furan-fused heterocycles are important substructures in many natural and synthetic bioactive compounds and can often serve as "'privileged structures"' in medical and pharmaceutical chemistry.[1] Among them, tricyclic indenofurans appear as key structural motifs that exist in many natural products such as (−)-galiellalactone,[2] leucosceptroid A,[3] and ramelteon,[4] which display important biological activities. With these attributes in mind, a great deal of effort has been devoted to carrying out various synthetic strategies for accessing indenofurans and their structural analogues.[5] However, there are only a few reports related to the synthesis of indeno[1,2-*c*]furans, which involved transannulation of *o*-lithiated aryloxiranes,[6] Pd-catalyzed bicyclization of 2-alkynyliodobenzenes,[7] and gold-catalyzed cyclization of aryl dienyl ketones[8a] or 1,6-dialkynols[8b] as well as other methods.[9] Nevertheless, most of these approaches suffered from prefunctionalization of substrates, metal catalysts, narrow substrate scope, and laborious workup. Therefore, the development of a new method that is metal-free, convenient to carry out, and efficient toward functional indeno[1,2-*c*]furans from simple substrates continues to be of great interest in the scientific community.

Multicomponent bicyclization reactions enable rapid access to structural variation and complexity within single-step conversions for the collection of highly functionalized polycyclic structures of chemical and pharmaceutical interest.[10] Such reactions feature annulation efficiency and extreme convergence while minimizing the generation of waste.[11] Of particular interest to us has been the exceptional reactivity of isocyanides, allowing their direct bicyclizations toward complex and diverse heterocyclic compounds.[12] Recently, Li and co-workers reported a three-component reaction of *o*-phthalalde-hydes (OPA), isocyanides, and amines, affording structurally diverse 1-carboxamido-isoindoles (Scheme 1a).[13] On the basis

Scheme 1. Bicyclizations toward Indeno[1,2-*c*]furans



of this study and our recent findings on isocyanide-enabled transformations,[14] we questioned whether the reaction selectivity could be harnessed to develop a new and challenging bicyclization cascade by controlling the reactivity of isocyanides and α-diazoketones in one pot due to the fact that donor−acceptor isocyanides[15] and α-diazoketones[16] inherently bear both nucleophilic and electrophilic character. Interestingly, mild base-promoted three-component reaction of *o*-phthalaldehydes (OPA), isocyanides, and α-diazoketones proceeded smoothly, enabling a direct and unprecedented [4 + 1]/[3 + 1 + 1] bicyclization cascade to access unexpected functionalized indeno[1,2-*c*]furans in a highly selective and functional-group-compatible manner (Scheme 1b). To the best of our knowledge, this metal-free three-component bicyclization for the construction of tricyclic indeno[1,2-*c*]furans via dediazotized C−H functionalization remains unexplored so far. Herein, we would like to elaborate this fascinating transformation using readily available OPA **1**, isocyanides **2**, and α-diazoketones **3**. This protocol represents the first domino procedure for the

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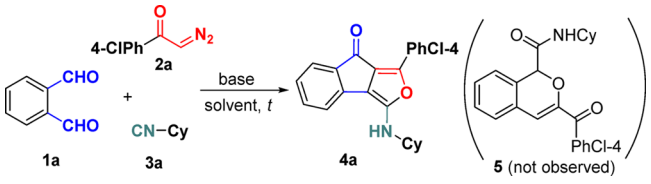
three-component synthesis of these new indeno[1,2-*c*]furans through a mild base-promoted metal-free dediazotized bicyclization, indicating that two different donor−acceptor isocyanides and α-diazoketones could be well compatible in the current reaction system.

RESULTS AND DISCUSSION

At the outset, we selected OPA (**1a**) with 1-(4-chlorophenyl)-2-diazoethanone (**2a**) and cyclohexyl isocyanide (**3a**) as benchmark substrates to investigate the feasibility of this three-component reaction. The reaction of **1a** with **2a** and **3a** was conducted in a 1.2:1.2:1 molar ratio in acetonitrile (CH_3CN) at 80 °C for 12 h, affording a red solid by column chromatography. After characterization by its spectroscopic data, it was interesting to find that the isolated product was a novel tricyclic indeno[1,2-*c*]furan **4a** rather than the expected isochromene **5**, albeit with a low 10% yield (Table 1, entry 1).

Table 1. Optimization of Reaction Conditions[a]



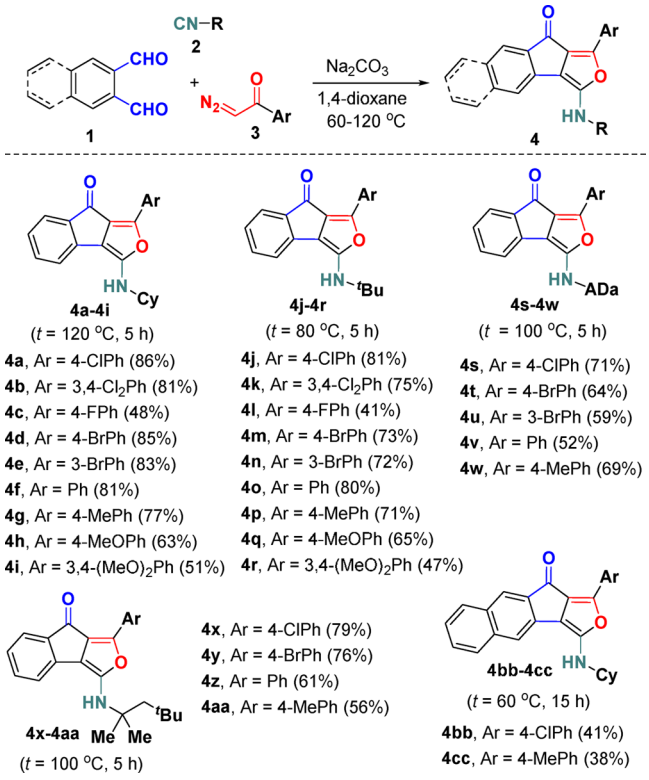
entry	base (equiv)	solvent	*T* (°C)	yield[b] (%)
1		CH_3CN	80	10
2	K_2CO_3 (1.0)	CH_3CN	80	17
3	Na_2CO_3 (1.0)	CH_3CN	80	24
4	Cs_2CO_3 (1.0)	CH_3CN	80	12
5	DBU (1.0)	CH_3CN	80	10
6	K_3PO_4 (1.0)	CH_3CN	80	<5
7	NaOAc (1.0)	CH_3CN	80	NR
8	Na_2CO_3 (1.0)	1,4-dioxane	80	29
9	Na_2CO_3 (1.0)	DMF	80	13
10	Na_2CO_3 (1.0)	DMSO	80	11
11	Na_2CO_3 (1.0)	EtOH	80	<5
12	Na_2CO_3 (1.0)	DCE	80	NR
13	Na_2CO_3(1.0)	toluene	80	<5
14	Na_2CO_3 (1.0)	1,4-dioxane	100	38
15	Na_2CO_3 (1.0)	1,4-dioxane	120	49
16	Na_2CO_3 (1.0)	1,4-dioxane	130	47
17	Na_2CO_3 (2.0)	1,4-dioxane	120	54
18	Na_2CO_3 (3.0)	1,4-dioxane	120	58
19	Na_2CO_3 (3.0)	1,4-dioxane	120	73[c]
20	Na_2CO_3 (3.0)	1,4-dioxane	120	86[d]

[a]Reaction conditions OPA (**1a**, 0.24 mmol), 1-(4-chlorophenyl)-2-diazoethanone (**2a**, 0.24 mmol), cyclohexyl isocyanide (**3a**, 0.2 mmol), solvent (2.0 mL). [b]Isolated yield based on substrate **3**. [c]Ratio of **1a**:**2a**:**3a** of 2:2:1. [d]Ratio of **1a**:**2a**:**3a** of 3:3:1.

This unexpected result prompted us to search for the optimal reaction conditions. The use of both K_2CO_3 and Na_2CO_3 as bases could improve this transformation (compare entry 1 and entries 2−3), and Na_2CO_3 gave the best outcome (24% yield, entry 3). Other bases (1.0 equiv), such as Cs_2CO_3, DBU, K_3PO_4, and NaOAc, all met with little success in three-component bicyclization (entries 4−7). Next, the effect of solvents was investigated. Exchanging acetonitrile (CH_3CN) for 1,4-dioxane led to a slightly higher yield of **4a** (entry 8). Other attempted solvents including *N*,*N*-dimethylformamide (DMF), dimethyl sulfoxide (DMSO), ethanol (EtOH), 1,2-dichloroethane (DCE), and toluene proved to be far less effective than 1,4-dioxane (entries 9−13 vs entry 8). It was also found that the reaction temperature exerted an important impact on the reaction efficiency. Higher conversion was observed with the reaction temperature at 120 °C (entries 14−16). Our next endeavor to change other reaction parameters was attempted to improve the yield of **4a**, including loading of Na_2CO_3 (1.0 to 3.0 equiv) and substrate ratio (entries 17−20). After careful optimization, we found that adjusting the substrate ratio to 3:3:1 (**1a**:**2a**:**3a**) in the presence of 3.0 equiv of Na_2CO_3 in 1,4-dioxane at 120 °C for 5.0 h further facilitated the reaction process and afforded 86% yield of **4a** (entry 20).

With the established optimal conditions, we then set out to explore the generality of this metal-free bicyclization with respect to a variety of isocyanides **2** and α-diazoketones **3** (Scheme 2). Those substituents on the phenyl ring of both isocyanides **2** and α-diazoketones **3** were proven not to hamper this domino cyclization, and a wide range of new functionalized indeno[1,2-*c*]furans **4a**−**4aa** can be obtained in moderate to good yields and in a functional-group-compatible fashion. Upon repeating the reaction with **1a** and **2a**, substrates **3** possessing either electronically neutral, poor, or rich substituents on the phenyl ring all work well, delivering the corresponding polyfunctionalized indeno[1,2-*c*]furans **4a**−**4i** with yields ranging from 48 to 86%. Various functional groups including chloro, fluoro, bromo, methyl, and methoxy are well compatible. In light of these results, we considered varying the substituents of the isocyanides and selected *tert*-butyl (**2b**), adamantyl (Ada, **2c**), and 1,1,3,3-tetramethylbutyl (**2d**)

Scheme 2. Substrate Scope for Synthesis of 4[a]



[a]Reaction conditions **1** (0.6 mmol), **2** (0.6 mmol) and **3** (0.2 mmol), 1,4-dioxane (2.0 mL), at 60−120 °C under air conditions. Yields of isolated products based on substrate **3** after column chromatography on silica gel are given.

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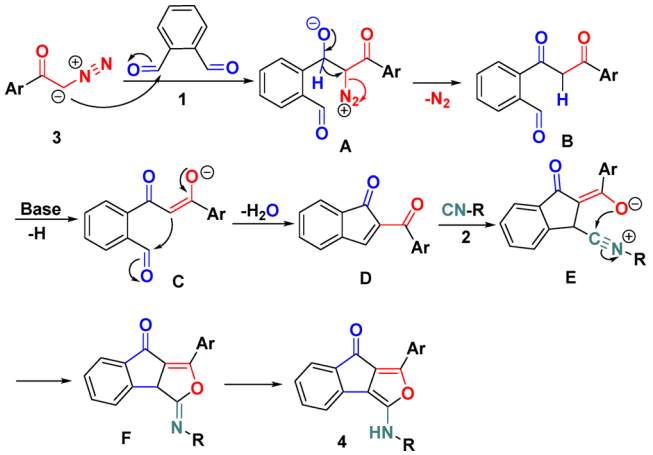
counterparts as three examples of representative isocyanides to evaluate the feasibility of the metal-free bicyclization reaction. As we had expected, all the isocyanides successfully participated in these transformations by suitably adjusting the reaction temperatures. For instance, *tert*-butyl isocyanide (**2b**) was subjected to the reaction of **1a** with **3** at 80 °C for 5 h, providing the desired indeno[1,2-*c*]furans **4j**−**4r** in 41−81% yields, whereas sterically encumbered isocyanide **2c** or **2d** was found to be successfully engaged in this α-diazoketone-enabled bicyclization with a reaction temperature of 100 °C. Alternatively, naphthalene-2,3-dicarbaldehyde **1b** was proven to be a suitable substrate, which underwent a similar three-component bicyclization process toward tetracyclic benzo[5,6]-indeno[1,2-*c*]furans **4bb** and **4cc** by lowering the reaction temperature and prolonging the reaction time, albeit with relatively low yields (38−41%). Obviously, the present bicyclizations can tolerate structurally diverse substrates with steric bulk and a different electronic nature, which offers a direct and practical protocol toward richly decorated indeno-[1,2-*c*]furans.

The structures of the resulting indeno[1,2-*c*]furans **4** were confirmed by carrying out their NMR and HRMS analysis. Furthermore, in the case of product **4g**, its structure was unambiguously determined using X-ray diffraction (see Supporting Information).

On the basis of the above experimental results and a literature survey,[16] a reasonable mechanism for forming products **4** is depicted in Scheme 3. First, the nucleophilic addition of α-diazoketone into one C═O group of OPA gives intermediates **A**, which undergo 1,2-hydride shift with concomitant loss of N_2[16e−g] to yield 1,3-dicarbonyl intermediates **B**.[17] Next, intermediates **B** are converted into 1-indenones **D** through 5-*exo-trig* cyclization and dehydration (Knoevenagel condensation-cyclization). Next, 1,4-addition of isocyanides **2** to 1-indenones **D** offers intermediates **E**, followed by oxo-5-*exo-dig* cyclization and subsequent tautomerization to form the final indeno[1,2-*c*]furans **4**.

Scheme 3. Proposed Mechanism for Forming 4



In conclusion, we have established an unprecedented, metal-free three-component [4 + 1]/[3 + 1 + 1] bicyclization of *o*-phthalaldehydes (OPA) with α-diazoketones and isocyanides that offered a facile and practical pathway to access a wide range of densely functionalized indeno[1,2-*c*]furans in a highly convergent manner. The present transformation involved an aldol-type addition, 5-*exo-trig* cyclization, and a 1,4-addition as well as oxo-5-*exo-dig* cyclization sequence, resulting in the formation of two new rings and four new chemical bonds including one C−O bond. This protocol also features accessibility of reaction substrates, flexible structural modification, and mild reaction conditions. A further investigation on evaluating the biological interest of these compounds is underway in our lab.

EXPERIMENTAL SECTION

General Procedure for the Synthesis of 4. *Example for the Synthesis of 4a.* A solution of phthalaldehyde **1a** (0.6 mmol, 3.0 equiv), cyclohexyl isonitrile **3a** (0.2 mmol, 1.0 equiv), 1-(4-chlorophenyl)-2-diazoethanone **2a** (0.6 mmol, 3.0 equiv), and Na_2CO_3 (0.6 mmol, 3.0 equiv) in 1,4-dioxane was stirred at 120 °C under air atmosphere for 5.0 h. After completion of the reaction as detected by TLC, the solvent was removed under vacuum. The residue was separated by column chromatography on silica gel (eluent, petroleum ether/ethyl acetate in 15:1 ratio (V/V)) to afford the pure red solid **4a**.

*1-(4-Chlorophenyl)-3-(cyclohexylamino)-8H-indeno[1,2-c]furan-8-one (**4a**).* *T* = 120 °C, red solid, 64.9 mg, 86% yield; mp 176−177 °C; IR (KBr, *ν*, cm^{-1}) 3326, 2930, 1659, 1627, 1490, 1089, 880, 829, 752; ^1H NMR (400 MHz, DMSO-d_6; *δ*, ppm) 8.12−8.04 (m, 2H), 7.61−7.55 (m, 3H), 7.54−7.48 (m, 2H), 7.31 (d, *J* = 8.0 Hz, 1H), 7.15−7.08 (m, 1H), 3.63−3.52 (m, 1H), 2.07−1.76 (m, 4H), 1.68−1.61 (m, 1H), 1.48−1.32 (m, 4H), 1.25−1.15 (m, 1H); ^{13}C NMR (100 MHz, DMSO-d_6; *δ*, ppm) 185.0, 149.3, 142.6, 139.8, 138.2, 134.9, 133.7, 129.6, 127.4, 126.1, 125.4, 124.9, 124.5, 121.7, 98.2, 52.7, 33.8, 25.7, 24.9; HRMS (APCI-TOF) *m*/*z* calcd for $C_{23}H_{21}ClNO_2^+$ 378.1261 $[M + H]^+$, found 378.1264.

*3-(Cyclohexylamino)-1-(3,4-dichlorophenyl)-8H-indeno[1,2-c]-furan-8-one (**4b**).* *T* = 120 °C, red solid, 66.6 mg, 81% yield; mp 165−166 °C; IR (KBr, *ν*, cm^{-1}) 3399, 2929, 1678, 1630, 1465, 1133, 886, 756, 723, 675; ^1H NMR (400 MHz, DMSO-d_6; *δ*, ppm) 8.22 (s, 1H), 7.87 (d, *J* = 8.8 Hz 1H), 7.68 (d, *J* = 8.8 Hz, 1H), 7.52 (d, *J* = 7.6 Hz, 1H), 7.50−7.43 (m, 2H), 7.33 (d, *J* = 8.0 Hz, 1H), 7.09 (t, *J* = 7.2 Hz, 1H), 3.58−3.47 (m, 1H), 2.16−1.69 (m, 4H), 1.68−1.60 (m, 1H), 1.50−1.29 (m, 4H), 1.25−1.16 (m, 1H); ^{13}C NMR (100 MHz, DMSO-d_6; *δ*, ppm) 185.0, 149.5, 140.7, 139.8, 138.0, 134.9, 132.2, 131.6, 131.1, 128.8, 126.3, 125.5, 124.9, 124.5, 123.8, 121.6, 98.5, 52.6, 33.8, 25.6, 24.9; HRMS (APCI-TOF) *m*/*z* calcd for $C_{23}H_{20}Cl_2NO_2^+$ 412.0871 $[M + H]^+$, found 412.0870.

*3-(Cyclohexylamino)-1-(4-fluorophenyl)-8H-indeno[1,2-c]furan-8-one (**4c**).* *T* = 120 °C, red solid, 34.8 mg, 48% yield; mp 153−154 °C; IR (KBr, *ν*, cm^{-1}) 3340, 2924, 1636, 1618, 1505, 1232, 1158, 880, 837, 758; ^1H NMR (400 MHz, DMSO-d_6; *δ*, ppm) 8.23−8.10 (m, 2H), 7.58 (d, *J* = 7.6 Hz, 1H), 7.56−7.49 (m, 2H) 7.38 (t, *J* = 8.8 Hz, 2H), 7.25 (d, *J* = 8.0 Hz, 1H), 7.16−7.08 (m, 1H), 3.63−3.51 (m, 1H), 2.10−1.94 (m, 2H), 1.84−1.72 (m, 2H), 1.68−1.61 (m, 1H), 1.49−1.32 (m, 4H), 1.26−1.16 (m, 1H); ^{13}C NMR (100 MHz, DMSO-d_6; *δ*, ppm) 185.0, 162.7 (J_{CF} = 246.6 Hz), 149.1, 143.1, 139.8, 138.4, 134.8, 126.9 (J_{CF} = 8.4 Hz), 125.4 (J_{CF} = 3.0 Hz), 124.9, 124.5, 124.4, 124.4, 121.7, 116.7 (J_{CF} = 22.0 Hz), 97.9, 52.7, 33.8, 25.7, 24.9; HRMS (APCI-TOF) *m*/*z* calcd for $C_{23}H_{21}FNO_2^+$ 362.1556 $[M + H]^+$, found 362.1556.

*1-(4-Bromophenyl)-3-(cyclohexylamino)-8H-indeno[1,2-c]furan-8-one (**4d**).* *T* = 120 °C, red solid, 71.5 mg, 85% yield; mp 198−199 °C; IR (KBr, *ν*, cm^{-1}) 3325, 2929, 1659, 1625, 1488, 880, 826, 751, 723; ^1H NMR (400 MHz, DMSO-d_6; *δ*, ppm) 8.03 (d, *J* = 8.8 Hz, 2H), 7.73 (d, *J* = 8.8 Hz, 2H), 7.58 (d, *J* = 7.6 Hz, 1H), 7.55−7.50 (m, 2H), 7.32 (d, *J* = 8.4 Hz, 1H), 7.15−7.10 (m, 1H), 3.62−3.54 (m, 1H), 2.05−1.98 (m, 2H), 1.83−1.75 (m, 2H), 1.68−1.61 (m, 1H), 1.49−1.34 (m, 4H), 1.25−1.17 (m, 1H); ^{13}C NMR (100 MHz, DMSO-d_6; *δ*, ppm) 185.1, 149.3, 142.7, 139.8, 138.2, 135.0, 132.5, 127.7, 126.3, 125.5, 125.0, 124.6, 122.5, 121.7, 98.3, 52.7, 33.8, 25.7, 24.9; HRMS (APCI-TOF) *m*/*z* calcd for $C_{23}H_{21}BrNO_2^+$ 422.0756 $[M + H]^+$, found 422.0766.

*1-(3-Bromophenyl)-3-(cyclohexylamino)-8H-indeno[1,2-c]furan-8-one (**4e**).* *T* = 120 °C, red solid, 70.0 mg, 83% yield; mp 142−143

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°C; IR (KBr, ν, cm^{-1}) 3394, 2931, 1676, 1630, 1474, 1448, 882, 755, 722; ^1H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.35−8.29 (m, 1H), 8.02−7.98 (m, 1H), 7.58−7.55 (m, 2H), 7.52−7.48 (m, 2H), 7.48−7.44 (m, 1H), 7.35 (d, J = 8.0 Hz, 1H), 7.14−7.09 (m, 1H), 3.59−3.53 (m, 1H), 2.04−1.76 (m, 4H), 1.67−1.62 (m, 1H), 1.47−1.34 (m, 4H), 1.25−1.18 (m, 1H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.1, 149.5, 141.8, 139.9, 138.1, 135.0, 131.9, 131.6, 130.6, 126.7, 126.0, 124.9, 124.6, 123.0, 122.7, 121.7, 98.3, 52.6, 33.8, 25.7, 24.9; HRMS (APCI-TOF) m/z calcd for $C_{23}H_{21}BrNO_2^+$ 422.0755 $[M + H]^+$, found 422.0779.

3-(Cyclohexylamino)-1-phenyl-8H-indeno[1,2-c]furan-8-one (**4f**). T = 120 °C, red solid, 55.4 mg, 81% yield; mp 181−182 °C; IR (KBr, ν, cm^{-1}) 3272, 2929, 1665, 1629, 1564, 1493, 1343, 1311, 1196, 1107, 879, 760; ^1H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.14 (d, J = 7.6 Hz, 2H), 7.59 (d, J = 7.6 Hz, 1H), 7.57−7.48 (m, 4H), 7.44−7.38 (m, 1H), 7.25 (d, J = 8.0 Hz, 1H), 7.16−7.09 (m, 1H), 3.66−3.53 (m, 1H), 2.08−1.97 (m, 2H), 1.85−1.74 (m, 2H), 1.69−1.60 (m, 1H), 1.51−1.32 (m, 4H), 1.27−1.18 (m, 1H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.0, 149.1, 144.0, 139.8, 138.4, 134.8, 129.7, 129.5, 128.6, 124.9, 124.8, 124.6, 124.5, 121.7, 98.1, 52.7, 33.8, 25.7, 25.0; HRMS (APCI-TOF) m/z calcd for $C_{23}H_{22}NO_2^+$ 344.1650 $[M + H]^+$, found 344.1633.

3-(Cyclohexylamino)-1-(p-tolyl)-8H-indeno[1,2-c]furan-8-one (**4g**). T = 120 °C, red solid, 55.1 mg, 77% yield; mp 175−176 °C; IR (KBr, ν, cm^{-1}) 3301, 2930, 1629, 1609, 1559, 1509, 1448, 1312, 881; ^1H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.04 (d, J = 8.4 Hz, 2H), 7.58 (d, J = 7.6 Hz, 1H), 7.55−7.49 (m, 2H), 7.34 (d, J = 8.0 Hz, 2H), 7.23 (d, J = 8.0 Hz, 1H), 7.14−7.09 (m, 1H), 3.61−3.52 (m, 1H), 2.36 (s, 3H), 2.06−1.98 (m, 2H), 1.83−1.75 (m, 2H), 1.68−1.61 (m, 1H), 1.48−1.33 (m, 4H), 1.25−1.17 (m, 1H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 184.8, 148.9, 144.5, 139.7, 138.6, 134.6, 130.0, 126.1, 124.8, 124.7, 124.4, 124.1, 121.7, 97.9, 52.7, 33.8, 25.7, 24.9, 21.6; HRMS (APCI-TOF) m/z calcd for $C_{24}H_{24}NO_2^+$ 358.1807 $[M + H]^+$, found 358.1815.

3-(Cyclohexylamino)-1-(4-methoxyphenyl)-8H-indeno[1,2-c]-furan-8-one (**4h**). T = 120 °C, red solid, 47.1 mg, 63% yield; mp 165−166 °C; IR (KBr, ν, cm^{-1}) 3295, 2927, 1634, 1607, 1563, 1509, 1255, 1178, 880, 832, 753; ^1H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.16−8.08 (m, 2H), 7.58 (d, J = 7.6 Hz, 1H), 7.55−7.48 (m, 2H), 7.17 (d, J = 8.0 Hz, 1H), 7.14−7.08 (m, 3H), 3.84 (s, 3H), 3.60−3.52 (m, 1H), 2.06−1.75 (m, 4H), 1.67−1.61 (m, 1H), 1.48−1.32 (m, 4H), 1.25−1.16 (m, 1H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 184.6, 160.7, 148.6, 144.9, 139.6, 138.8, 134.5, 126.6, 124.7, 124.3, 123.0, 121.7, 121.6, 115.0, 97.5, 55.8, 52.7, 33.8, 25.7, 24.9; HRMS (APCI-TOF) m/z calcd for $C_{24}H_{24}NO_3^+$ 374.1756 $[M + H]^+$, found 374.1742.

3-(Cyclohexylamino)-1-(3,4-dimethoxyphenyl)-8H-indeno[1,2-c]-furan-8-one (**4i**). T = 120 °C, red solid, 41.2 mg, 51% yield; mp 91−92 °C; IR (KBr, ν, cm^{-1}) 3326, 2927, 1667, 1631, 1565, 1516, 1465, 1254, 1222, 1114, 1024, 896, 754; ^1H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.01 (d, J = 1.6 Hz, 1H), 7.63−7.57 (m, 2H), 7.52−7.47 (m, 2H), 7.21 (d, J = 8.0 Hz, 1H), 7.15−7.10 (m, 2H), 3.89 (s, 3H), 3.84 (s, 3H), 3.60−3.52 (m, 1H), 2.06−1.99 (m, 2H), 1.83−1.75 (m, 2H), 1.68−1.62 (m, 1H), 1.47−1.34 (m, 4H), 1.25−1.16 (m, 1H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 184.6, 150.5, 149.4, 148.7, 145.1, 139.5, 138.8, 134.5, 124.8, 124.4, 123.2, 121.7, 121.6, 117.8, 112.4, 108.5, 97.4, 56.1, 55.9, 52.7, 33.8, 25.7, 24.9; HRMS (APCI-TOF) m/z calcd for $C_{25}H_{26}NO_4^+$ 404.1862 $[M + H]^+$, found 404.1888.

3-(tert-Butylamino)-1-(4-chlorophenyl)-8H-indeno[1,2-c]furan-8-one (**4j**). T = 80 °C, red solid, 57.1 mg, 81% yield; mp 148−149 °C; IR (KBr, ν, cm^{-1}) 3399, 2963, 1690, 1588, 1487, 1366, 1248, 1090, 849, 751; ^1H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.10 (d, J = 8.4 Hz, 2H), 7.82 (d, J = 7.6 Hz, 1H), 7.64−7.57 (m, 3H), 7.56−7.51 (m, 1H), 7.18−7.14 (m, 1H), 6.68 (s, 1H), 1.44 (s, 9H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.2, 148.9, 143.8, 139.6, 138.5, 134.8, 134.0, 129.8, 127.5, 126.2, 125.3, 124.8, 124.5, 122.4, 104.2, 53.6, 30.5; HRMS (ESI-TOF) m/z calcd for $C_{21}H_{17}ClNO_2^-$ 350.0948 $[M - H]^-$, found 350.0959.

3-(tert-Butylamino)-1-(3,4-dichlorophenyl)-8H-indeno[1,2-c]-furan-8-one (**4k**). T = 80 °C, red solid, 57.8 mg, 75% yield; mp 131−132 °C; IR (KBr, ν, cm^{-1}) 3266, 2973, 1697, 1544, 1467, 1391, 1190, 1137, 1027, 881, 827, 760; ^1H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.34 (d, J = 1.6 Hz, 1H), 7.93−7.90 (m, 1H), 7.84−7.79 (m, 2H), 7.60−7.53 (m, 2H), 7.19−7.14 (m, 1H), 6.83 (s, 1H), 1.44 (s, 9H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.2, 149.1, 143.0, 139.7, 138.4, 134.9, 132.1, 131.8, 130.7, 126.9, 125.4, 125.1, 124.8, 123.1, 122.9, 122.4, 104.2, 53.6, 30.5; HRMS (ESI-TOF) m/z calcd for $C_{21}H_{16}Cl_2NO_2^-$ 384.0558 $[M - H]^-$, found 384.0569.

3-(tert-Butylamino)-1-(4-fluorophenyl)-8H-indeno[1,2-c]furan-8-one (**4l**). T = 80 °C, red solid, 27.6 mg, yield 41%; mp 110−111 °C; IR (KBr, ν, cm^{-1}) 3434, 2970, 1677, 1648, 1503, 1220, 1157, 877, 828, 763; ^1H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.20−8.14 (m, 2H), 7.82 (d, J = 7.6 Hz, 1H), 7.59 (d, J = 7.6 Hz, 1H), 7.56−7.51 (m, 1H), 7.45−7.38 (m, 2H), 7.20−7.14 (m, 1H), 6.59 (s, 1H), 1.43 (s, 9H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.06, 162.8 (J_{CF} = 246.6 Hz), 148.6, 144.4, 139.6, 138.7, 134.7, 127.0 (J_{CF} = 8.4 Hz), 125.5 (J_{CF} = 3.1 Hz), 125.3, 124.7, 123.6, 122.4, 116.8 (J_{CF} = 22.2 Hz), 104.4, 53.6, 30.6; HRMS (ESI-TOF) m/z calcd for $C_{21}H_{17}FNO_2^-$ 334.1243 $[M - H]^-$, found 334.1265.

1-(4-Bromophenyl)-3-(tert-butylamino)-8H-indeno[1,2-c]furan-8-one (**4m**). T = 80 °C, red solid, 57.9 mg, 73% yield; mp 92−93 °C; IR (KBr, ν, cm^{-1}) 3393, 2970, 1678, 1645, 1614, 1484, 1395, 1070, 1005, 878; ^1H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.06−8.00 (m, 2H), 7.83−7.81 (m, 1H), 7.77−7.74 (m, 2H), 7.58 (d, J = 7.6 Hz, 1H), 7.55−7.51 (m, 1H), 7.19−7.13 (m, 1H), 6.69 (s, 1H), 1.44 (s, 9H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.2, 148.9, 143.9, 139.6, 138.5, 134.8, 132.6, 127.8, 126.4, 125.3, 124.8, 124.6, 122.7, 122.4, 104.3, 53.6, 30.5; HRMS (ESI-TOF) m/z calcd for $C_{21}H_{17}BrNO_2^-$ 394.0443 $[M - H]^-$, found 394.0470.

1-(3-Bromophenyl)-3-(tert-butylamino)-8H-indeno[1,2-c]furan-8-one (**4n**). T = 80 °C, red solid, 57.2 mg, 72% yield; mp 127−128 °C; IR (KBr, ν, cm^{-1}) 3493, 2989, 1679, 1675, 1654, 1489, 1396, 1078, 1045, 876, 838; ^1H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.37−8.33 (m, 1H), 7.98 (d, J = 2.0 Hz, 1H), 7.82 (d, J = 7.6 Hz, 1H), 7.61−7.56 (m, 2H), 7.56−7.46 (m, 2H), 7.19−7.15 (m, 1H), 6.76 (s, 1H), 1.44 (s, 9H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.2, 149.1, 143.0, 139.7, 138.4, 134.9, 132.1, 131.8, 130.7, 126.9, 125.4, 125.1, 124.8, 123.1, 122.9, 122.4, 104.2, 53.6, 30.5; HRMS (ESI-TOF) m/z calcd for $C_{21}H_{17}BrNO_2^-$ 394.0443 $[M - H]^-$, found 394.0468.

3-(tert-Butylamino)-1-phenyl-8H-indeno[1,2-c]furan-8-one (**4o**). T = 80 °C, red solid, 50.5 mg, 80% yield; mp 107−108 °C; IR (KBr, ν, cm^{-1}) 3338, 2978, 1756, 1695, 1615, 1469, 1396, 1343, 1180, 829, 765; ^1H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.15−8.10 (m, 2H), 7.82 (d, J = 8.0 Hz, 1H), 7.59 (d, J = 7.6 Hz, 1H), 7.57−7.51 (m, 3H), 7.46−7.41 (m, 1H), 7.20−7.14 (m, 1H), 6.62 (s, 1H), 1.44 (s, 9H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.1, 148.6, 145.2, 139.6, 138.7, 134.6, 129.9, 129.6, 128.7, 125.3, 124.7, 124.0, 122.3, 104.4, 53.6, 30.5; HRMS (ESI-TOF) m/z calcd for $C_{21}H_{18}NO_2^-$ 316.1338 $[M - H]^-$ found 316.1336.

3-(tert-Butylamino)-1-(p-tolyl)-8H-indeno[1,2-c]furan-8-one (**4p**). T = 80 °C, red solid, 47.2 mg, 71% yield; mp 82−83 °C; IR (KBr, ν, cm^{-1}) 3318, 2968, 1736, 1675, 1605, 1462, 1366, 1313, 1183, 826, 755; ^1H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.04 (d, J = 8.0 Hz, 2H), 7.82 (d, J = 7.6 Hz, 1H), 7.59 (d, J = 7.6 Hz, 1H), 7.55−7.50 (m, 1H), 7.36 (d, J = 8.0 Hz, 2H), 7.16 (t, J = 7.6 Hz, 1H), 6.52 (s, 1H), 2.37 (s, 3H), 1.43 (s, 9H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 184.9, 148.3, 145.8, 139.9, 139.5, 138.9, 134.5, 130.1, 126.2, 125.2, 124.8, 124.6, 123.2, 122.3, 104.5, 53.5, 30.5, 21.6; HRMS (ESI-TOF) m/z calcd for $C_{22}H_{20}NO_2^-$ 330.1494 $[M - H]^-$, found 330.1500.

3-(tert-Butylamino)-1-(4-methoxyphenyl)-8H-indeno[1,2-c]-furan-8-one (**4q**). T = 80 °C, red solid, 45.3 mg, 65% yield; mp 167−168 °C; IR (KBr, ν, cm^{-1}) 3336, 2966, 1668, 1651, 1607, 1506, 1258, 1176, 1020, 877, 838, 736; ^1H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.16−8.05 (m, 2H), 7.81 (d, J = 7.6 Hz, 1H), 7.59 (d, J = 7.6 Hz, 1H), 7.55−7.49 (m, 1H), 7.20−7.10 (m, 3H), 6.45 (s, 1H), 3.84 (s, 3H), 1.42 (s, 9H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 184.7, 160.9, 148.0, 146.2, 139.3, 139.1, 134.3, 126.8, 125.2, 124.5, 122.4, 122.0, 121.7, 115.2, 104.8, 55.9, 53.6, 30.6; HRMS (ESI-TOF) m/z calcd for $C_{22}H_{20}NO_3^-$ 346.1443 $[M - H]^-$, found 346.1471.

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*3-(tert-Butylamino)-1-(3,4-dimethoxyphenyl)-8H-indeno[1,2-c]-furan-8-one (**4r**).* T = 80 °C, red solid, 35.6 mg, 47% yield; mp 232−233 °C; IR (KBr, ν, cm^{-1}) 3277, 2929, 1647, 1632, 1560, 1459, 1262, 1140, 1028, 866, 793; ^{1}H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.02 (d, J = 2.0 Hz, 1H), 7.82 (d, J = 7.6 Hz, 1H), 7.60−7.55 (m, 2H), 7.54−7.50 (m, 1H), 7.20−7.13 (m, 2H), 6.47 (s, 1H), 3.90 (s, 3H), 3.84 (s, 3H), 1.42 (s, 9H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 184.8, 150.6, 149.4, 148.1, 146.4, 139.3, 139.1, 134.3, 125.2, 124.5, 122.4, 122.2, 121.7, 117.7, 112.5, 108.7, 104.7, 56.1, 55.9, 53.6, 30.5; HRMS (ESI-TOF) m/z calcd for $C_{23}H_{22}NO_4^-$ 376.1549 [M − H]$^-$, found 376.1555.

*3-(Adamantan-1-ylamino)-1-(4-chlorophenyl)-8H-indeno[1,2-c]-furan-8-one (**4s**).* T = 100 °C, red solid, 61.1 mg, 71% yield; mp 190−191 °C; IR (KBr, ν, cm^{-1}) 3445, 2904, 1690, 1612, 1487, 1089, 879, 834, 727; ^{1}H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.13−8.07 (m, 2H), 7.82 (d, J = 7.6 Hz, 1H), 7.66−7.61 (m, 2H), 7.59 (d, J = 7.2 Hz, 1H), 7.56−7.52 (m, 1H), 7.20−7.15 (m, 1H), 6.54 (s, 1H), 2.12 (s, 3H, CH), 2.00 (d, J = 2.4 Hz, 6H), 1.68 (s, 6H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.1, 148.4, 144.1, 139.5, 138.7, 134.8, 134.1, 130.9, 129.8, 127.5, 126.3, 125.5, 124.8, 124.3, 122.5, 54.2, 43.3, 36.3, 29.6; HRMS (ESI-TOF) m/z calcd for $C_{27}H_{23}ClNO_2^-$ 428.1417 [M − H]$^-$, found 428.1419.

*3-(Adamantan-1-ylamino)-1-(4-bromophenyl)-8H-indeno[1,2-c]-furan-8-one (**4t**).* T = 100 °C, red solid, 60.8 mg, 64% yield; mp 197−198 °C; IR (KBr, ν, cm^{-1}) 3265, 2903, 1695, 1557, 1485, 1357, 1303, 1188, 1089, 1006, 878, 831, 726; ^{1}H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.05−8.01 (m, 2H), 7.82 (d, J = 7.6 Hz, 1H), 7.77 (d, J = 8.4 Hz, 2H), 7.59 (d, J = 7.6 Hz, 1H), 7.57−7.52 (m, 1H), 7.18 (t, J = 7.6 Hz, 1H), 6.54 (s, 1H), 2.12 (s, 3H, CH), 2.03−1.96 (m, 6H), 1.68 (s, 6H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.1, 148.4, 144.2, 139.5, 138.6, 134.8, 132.7, 127.8, 126.5, 125.6, 124.8, 124.5, 122.9, 122.5, 105.8, 54.2, 43.3, 36.3, 29.6; HRMS (ESI-TOF) m/z calcd for $C_{27}H_{23}BrNO_2^-$ 472.0912 [M − H]$^-$, found 472.0931.

*3-(Adamantan-1-ylamino)-1-(3-bromophenyl)-8H-indeno[1,2-c]-furan-8-one (**4u**).* T = 100 °C, red solid, 56.0 mg, 59% yield; mp 178−179 °C; IR (KBr, ν, cm^{-1}) 3419, 2903, 1690, 1647, 1614, 1587, 1550, 1471, 1306, 858, 722; ^{1}H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.36 (s, 1H), 7.97 (d, J = 7.6 Hz, 1H), 7.82 (d, J = 7.6 Hz, 1H), 7.63−7.58 (m, 2H), 7.57−7.47 (m, 2H), 7.22−7.14 (m, 1H), 2.12 (s, 3H, CH), 2.01 (s, 6H), 1.69 (s, 6H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.2, 148.6, 143.2, 139.6, 138.5, 134.9, 132.2, 131. 9, 130.8, 127.1, 125.5, 125.0, 124.8, 123.1, 122.9, 122.5, 105.4, 54.2, 43.2, 36.3, 29.6; HRMS (ESI-TOF) m/z calcd for $C_{27}H_{23}BrNO_2^-$ 472.0912 [M − H]$^-$, found 472.0932.

*3-(Adamantan-1-ylamino)-1-phenyl-8H-indeno[1,2-c]furan-8-one (**4v**).* T = 100 °C, red solid, 41.2 mg, 52% yield; mp 145−146 °C; IR (KBr, ν, cm^{-1}) 3445, 2904, 1690, 1612, 1487, 1089, 879, 834, 727; ^{1}H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.03−7.98 (m, 2H), 7.69 (d, J = 7.6 Hz, 1H), 7.49−7.40 (m, 4H), 7.33−7.28 (m, 1H), 7.07−7.02 (m, 1H), 6.32 (s, 1H), 1.97 (s, 3H, CH), 1.89−1.86 (m, 6H), 1.54 (s, 6H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.1, 148.1, 145.5, 139.5, 138.9, 134.6, 129.9, 129.6, 128.7, 125.5, 124.8, 124.7, 123.8, 122.4, 105.9, 54.1, 43.3, 36.3, 29.6; HRMS (ESI-TOF) m/z calcd for $C_{27}H_{24}NO_2^-$ 394.1807 [M − H]$^-$, found 394.1831.

*3-(Adamantan-1-ylamino)-1-(p-tolyl)-8H-indeno[1,2-c]furan-8-one (**4w**).* T = 100 °C, red solid, 56.3 mg, 69% yield; mp 157−158 °C; IR (KBr, ν, cm^{-1}) 3288, 2905, 1675, 1611, 1508, 1308, 877, 768, 740; ^{1}H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.03 (d, J = 8.0 Hz, 2H), 7.81 (d, J = 7.6 Hz, 1H), 7.58 (d, J = 7.6 Hz, 1H), 7.56−7.51 (m, 1H), 7.37 (d, J = 8.0 Hz, 2H), 7.20−7.15 (m, 1H), 6.35 (s, 1H), 2.37 (s, 3H), 2.11 (s, 3H), 2.02−1.93 (m, 6H), 1.67 (s, 6H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 184.9, 147.8, 146.1, 140.0, 139.3, 139.1, 134.5, 130.2, 126.2, 125.5, 124.9, 124.6, 123.0, 122.4, 106.3, 54.1, 43.4, 36.3, 29.6, 21.7; HRMS (ESI-TOF) m/z calcd for $C_{28}H_{26}NO_2^-$ 408.1964 [M − H]$^-$, found 408.1982.

*1-(4-Chlorophenyl)-3-((2,4,4-trimethylpentan-2-yl)amino)-8H-indeno[1,2-c]furan-8-one (**4x**).* T = 100 °C, red solid, 64.5 mg, 79% yield; mp 110−111 °C; IR (KBr, ν, cm^{-1}) 3333, 2946, 1695, 1614, 1488, 1217, 1089, 878, 833, 761; ^{1}H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.12−8.07 (m, 2H), 7.83 (d, J = 7.6 Hz, 1H), 7.63 (d, J = 8.8 Hz, 2H), 7.57 (d, J = 7.2 Hz, 1H), 7.55−7.49 (m, 1H), 7.16−7.11 (m, 1H), 6.73 (s, 1H), 1.80 (s, 2H), 1.49 (s, 6H), 1.01 (s, 9H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.2, 149.0, 143.4, 139.8, 138.3, 134.7, 133.8, 129.8, 127.5, 126.0, 124.9, 124.8, 124.7, 122.3, 102.7, 57.0, 52.9, 31.9, 31.7, 31.2; HRMS (APCI-TOF) m/z calcd for $C_{25}H_{27}ClNO_2^+$ 408.1730 [M + H]$^+$, found 408.1724.

*1-(4-Bromophenyl)-3-((2,4,4-trimethylpentan-2-yl)amino)-8H-indeno[1,2-c]furan-8-one (**4y**).* T = 100 °C, red solid, 68.9 mg, 76% yield; mp 96−97 °C; IR (KBr, ν, cm^{-1}) 3270, 2952, 1694, 1569, 1485, 1362, 1218, 1006, 877, 830, 752; ^{1}H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.02 (d, J = 8.8 Hz, 2H), 7.83 (d, J = 7.6 Hz, 1H), 7.76 (d, J = 8.8 Hz, 2H), 7.57 (d, J = 7.6 Hz, 1H), 7.53 (t, J = 7.6 Hz, 1H), 7.13 (t, J = 7.6 Hz, 1H), 6.75 (s, 1H), 1.80 (s, 2H), 1.50 (s, 6H), 1.01 (s, 9H). ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.2, 149.0, 143.4, 139.8, 138.2, 134.7, 132.7, 127.8, 126.1, 124.9, 124.8, 124.7, 122.5, 122.3, 102.6, 57.0, 52.9, 31.9, 31.7, 31.2; HRMS (APCI-TOF) m/z calcd for $C_{25}H_{27}BrNO_2^+$ 452.1225 [M + H]$^+$, found 452.1241.

*1-Phenyl-3-((2,4,4-trimethylpentan-2-yl)amino)-8H-indeno[1,2-c]furan-8-one (**4z**).* T = 100 °C, red solid, 45.6 mg, 61% yield; mp 76−77 °C; IR (KBr, ν, cm^{-1}) 3329, 2979, 1678, 1663, 1613, 1489, 1222, 1192, 876, 755, 725; ^{1}H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.16−8.09 (m, 2H), 7.83 (d, J = 8.0 Hz, 1H), 7.59−7.50 (m, 4H), 7.45−7.39 (m, 1H), 7.18−7.10 (m, 1H), 6.67 (s, 1H), 1.81 (s, 2H), 1.50 (s, 6H), 1.01 (s, 9H). ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.2, 148.7, 144.7, 139.8, 138.4, 134.6, 129.7, 129.6, 128.8, 124.9, 124.7, 124.5, 124.2, 122.2, 102.7, 56.9, 52.9, 31.9, 31.7, 31.2; HRMS (APCI-TOF) m/z calcd for $C_{25}H_{28}NO_2^+$ 374.2120 [M + H]$^+$, found 374.2142.

*1-(p-Tolyl)-3-((2,4,4-trimethylpentan-2-yl)amino)-8H-indeno[1,2-c]furan-8-one (**4aa**).* T = 100 °C, red solid, 43.4 mg, 56% yield; mp 132−133 °C; IR (KBr, ν, cm^{-1}) 3341, 2950, 1673, 1611, 1506, 1468, 1311, 1222, 1142, 992, 876, 836, 75; ^{1}H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.03 (d, J = 8.0 Hz, 2H), 7.83 (d, J = 7.6 Hz, 1H), 7.57 (d, J = 7.2 Hz, 1H), 7.51 (t, J = 7.6 Hz, 1H), 7.36 (d, J = 8.0 Hz, 2H), 7.13 (t, J = 7.6 Hz, 1H), 6.61 (s, 1H), 2.37 (s, 3H), 1.80 (s, 2H), 1.49 (s, 6H), 1.01 (s, 9H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.0, 148.4, 145.3, 139.7, 139.7, 138.6, 134.4, 130.2, 126.2, 124.8, 124.6, 124.6, 123.4, 122.2, 102.6, 56.9, 52.9, 31.9, 31.7, 31.2, 21.6; HRMS (APCI-TOF) m/z calcd for $C_{26}H_{30}NO_2^+$ 388.2277 [M + H]$^+$, found 388.2289.

*1-(4-Chlorophenyl)-3-(cyclohexylamino)-10H-benzo[5,6]indeno-[1,2-c]furan-10-one (**4bb**).* T = 60 °C, red solid, 35 mg, 41% yield; mp 196−197 °C; IR (KBr, ν, cm^{-1}) 3271, 2931, 1682, 1625, 1487, 1168, 1088, 1011, 832, 789, 745; ^{1}H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.22 (s, 1H), 8.10 (d, J = 8.8 Hz, 2H), 8.02 (d, J = 8.0 Hz, 1H), 7.88 (s, 1H), 7.81 (d, J = 8.0 Hz, 1H), 7.62−7.53 (m, 3H), 7.46−7.36 (m, 2H), 3.72−3.63 (m, 1H), 2.09 (s, 1H), 2.06 (s, 1H), 1.87−1.76 (m, 2H), 1.68−1.66 (m, 1H), 1.56−1.38 (m, 4H), 1.28−1.20 (m, 1H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.2, 149.7, 141.5, 138.4, 137.2, 133.8, 133.3, 131.0, 130.7, 129.6, 129.4, 128.0, 127.7, 126.5, 126.0, 125.8, 125.7, 118.5, 99.5, 52.7, 34.0, 25.7, 25.1; HRMS (APCI-TOF) m/z calcd for $C_{27}H_{23}ClNO_2^+$ 428.1417 [M + H]$^+$, found 428.1451.

*3-(Cyclohexylamino)-1-(p-tolyl)-10H-benzo[5,6]indeno[1,2-c]-furan-10-one (**4cc**).* T = 60 °C, red solid, 30.9 mg, 38% yield; mp 145−146 °C; IR (KBr, ν, cm^{-1}) 3313, 2926, 1671, 1628, 1509, 1164, 869, 821, 784, 759; ^{1}H NMR (400 MHz, DMSO-d_6; δ, ppm) 8.21 (s, 1H), 8.10−7.97 (m, 3H), 7.88 (s, 1H), 7.82 (d, J = 8.0 Hz, 1H), 7.56 (t, J = 7.2 Hz, 1H), 7.42 (t, J = 7.2 Hz, 1H), 7.34 (d, J = 8.0 Hz, 2H), 7.28 (d, J = 8.0 Hz, 1H), 3.66 (s, 1H), 2.36 (s, 3H), 2.14−1.77 (m, 4H), 1.72−1.63 (m, 1H), 1.55−1.37 (m, 4H), 1.25−1.18 (m, 1H); ^{13}C NMR (100 MHz, DMSO-d_6; δ, ppm) 185.0, 149.2, 143.5, 139.2, 138.7, 137.1, 133.9, 130.9, 130.7, 130.0, 129.2, 128.0, 126.3, 125.7, 125.6, 125.2, 124.5, 118.5, 99.0, 52.8, 34.0, 25.8, 25.0, 21.6; HRMS (APCI-TOF) m/z calcd for $C_{28}H_{26}NO_2^+$ 408.1964 [M + H]$^+$, found 408.1979.

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■ ASSOCIATED CONTENT

Supporting Information

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^{1}H and ^{13}C NMR spectra for all pure products (PDF)
X-ray crystal data (CIF) for **4g** (CIF)

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Notes

The authors declare no competing financial interest.

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