Accepted Manuscript

Molecular diversity of cycloaddition reactions of the functionalized pyridinium salts with 3-phenacylideneoxindoles

Qin Fu, Chao-Guo Yan

PII: S0040-4020(13)00769-2

DOI: 10.1016/j.tet.2013.05.034

Reference: TET 24372

To appear in: *Tetrahedron*

Received Date: 28 February 2013

Revised Date: 26 April 2013

Accepted Date: 13 May 2013

Please cite this article as: Fu Q, Yan C-G, Molecular diversity of cycloaddition reactions of the functionalized pyridinium salts with 3-phenacylideneoxindoles, *Tetrahedron* (2013), doi: 10.1016/j.tet.2013.05.034.

Molecular diversity of cycloaddition reactions of the functionalized pyridinium salts with 3-phenacylideneoxindoles

Qin Fu, Chao-Guo Yan*

College of Chemistry & Chemical Engineering, Yangzhou University, Yangzhou 225002, China

Tel: 86-514-87975531; Fax: 86-514-87975244; e-mail: cgyan@yzu.edu.cn

Abstract: The cycloaddition reactions of a series of functionalized pyridinium salts, which were generated from reaction of pyridine with active alkyl halides such as *p*-nitrobenzyl bromide, *N*,*N*-diethyl chloroacetamide and phenacyl bromides with 3-phenacylideneoxindoles in the presence of triethylamine showed very interesting molecular diversities. A series of the functionalized spiro[cyclopropane-1,3'-indolines], and 3-furan-3(2H)-ylidene)indolin-2-ones were successfully prepared depending upon the structures of the pyridinium salts and reaction conditions. The regioselectivity and stereoselectivity of the reactions as well as reaction mechanisms were briefly discussed.

Keywords: heterocycle, spiro compound, pyridinium *N*-ylide, oxindole, cyclopropane.

1. Introduction

The spirooxindole unit is a privileged heterocyclic motif that forms the core structure of a large family of natural alkaloids and many pharmacological agents with important bioactivity and interesting structural properties.[1,2] The unique structures and the highly pronounced pharmacological activity displayed by the spirooxindoles have made them attractive synthetic targets.[3] In various heterocyclic and carbocyclic spirooxindoles, the spirocyclopentyl and spirocyclohexyl systems, especially spiro[indoline-pyrrolines] and spiro[indoline-pyridines] have

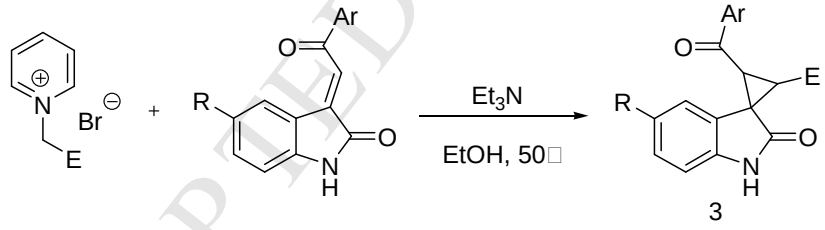
been deeply and extensively investigated.[4] On the other hand the highly strained spiro[cyclopropane-1,3'-indolines] have attracted limited attentions. Recently functionalized spiro[cyclopropane-1,3'-indolines] were discovered having inotropic herbicidal properties and as an HIV non-nucleoside reverse transcriptase inhibitors.[5] spiro[cyclopropane-1,3'-indolines] were also employed as building blocks for construction of more complex oxindole compounds.[6] As a consequence, a number of methods have been reported for the facile preparation of spiro[cyclopropane-1,3'-indolines]. The known preparation methods included cycloaddition of diazomethane with isatins or diazoamide with cyclic ketones,[7] cycloaddition of sulfur[8], phosphrous and arsonium[9] ylides with isatins, dialkylation of 3-acetonitriloxindole with alkyl dibromides,[10] reductive cyclization of Baylis–Hillman adducts of isatin and others.[11] An enantioselective nitrocycloproppanation of oxindoles through the direct reaction of bromonitroalkyl derivatives and Boc-protected 3-alkylidene oxindoles were also successfully developed.[12] Recently we developed an efficient synthetic procedure for the polysubstituted cyclopropanes via the domino reaction of *in situ* generated pyridinium salts with electron-deficient olefins.[13] Pyridinium salts have been used extensively in cycloadditions for the synthesis of the fused heterocycles with a nitrogen at the point of fusion[14,15] and also been successfully employed to prepare some spirooxindole systems.[17,18] We envisaged that spiro[cyclopropane-1,3'-indolines] would be synthesized by the similar reactions of *in situ* generated pyridinium salts with 3-alkylidene oxindoles. A literature survey showed that there has been one briefly report of formation of spiro[cyclopropane-1,3'-indolines] by the reactions of *N*-phenacylpyridinium salts with phenacylideneoxindoles, in which only few synthetic examples were provided.[19] With the aim of expanding our previous studies on the synthesis of spirooxindoles,[20] we decided to

systematically investigated the reactions of a series of pyridinium salts with 3-phenacylideneoxindoles and report the interesting molecular diversities of this domino reaction.

2. Results and Discussion

In our initial endeavour, according to our previously established reaction conditions for preparation of cyclopropanes,[13] a solution of eqular molar *p*-nitrobenzylpyridinium bromide (**1a**) and 3-phenacylideneoxindoles (**2a-2d**) in ethanol with triethylamine as base catalyst was carried out at elevated temperature for six hours. After workup, we were pleased to find that the expected pure functionalized spiro[cyclopropane-1,3'-indolines] (**3a-3d**) was obtained in 65-78% yields (Table 1, entries 1-4). It showed that this reaction is a facile protocol for synthesis of spiro[cyclopropane-1,3'-indolines] with advantages of mild reaction conditions, easily accessible starting material and easy purification of the products. Then under similar conditions the reactions

Table 1 Reactions of pyridinium salts with 3-phenacylideneoxindoles



3

Entry	Compd	E	R	Ar	Yield[a] (%)
1	**3a**	*p*-$NO_2C_6H_4$	H	C_6H_5	78
2	**3b**	*p*-$NO_2C_6H_4$	H	*p*-ClC_6H_4	76
3	**3c**	*p*-$NO_2C_6H_4$	F	C_6H_5	65
4	**3d**	*p*-$NO_2C_6H_4$	F	*p*-ClC_6H_4	70
5	**3e**	$CON(CH_2CH_3)_2$	CH_3	*p*-$CH_3C_6H_4$	72
6	**3f**	$CON(CH_2CH_3)_2$	CH_3	*p*-ClC_6H_4	88
7	**3g**	$CON(CH_2CH_3)_2$	F	C_6H_5	69
8	**3h**	$CON(CH_2CH_3)_2$	F	*p*-ClC_6H_4	54
9	**3i**	$CON(CH_2CH_3)_2$	Cl	*p*-ClC_6H_4	82
10	**3j**	$CON(CH_2CH_3)_2$	Cl	*p*-$CH_3C_6H_4$	80

a: isolate yields; b: pyridinium salt (1.1 mmol), 3-phenacylideneoxindole (1.0 mmol), triethylamine (0.2 mmol), EtOH (15 mL), 50°C, 6 h.

of *N*-diethylcarbamoylmethylpyridinium bromide (**1b**) with various 3-phenacylideneoxindoles also proceeded very smoothly to give the spiro[cyclopropane-1,3'-indolines] (**3e-3j**) in 54-88% yields (Table 1, entries 5-10). The substituents on oxindole moiety and aryl groups showed marginal effect on the yield of product.

The structures of the prepared spiro[cyclopropane-1,3'-indolines] were characterized with spectroscopic methods and confirmed by the X-ray diffraction determination of single crystal **3e** (**Figure 1**). In the ^{1}H NMR spectra of spiro compounds **3a-3j**, the two protons at 2,3-position of cyclopropyl ring usually display two doublets at about 4.30 and 4.10 ppm with the vicinal coupling constant J = 7.8Hz. It has been well known that in *trans*-cyclopropane the vicinal coupling constant of the two methine protons J = 7-10Hz, while in *cis*-cyclopropane the vicinal coupling constant J = 4-7Hz.[21] From **Figure 1** It can be seen clearly that the aroxyl group and diethylcarbamoyl group are in *trans*-configuration. On the other hand the aroxyl group and the phenyl group of oxindole moiety also exist in *trans*-position. It is well known that these two groups are in *cis*-position in starting 3-phenacylideneoxindoles.[22] This result undoubtedly showed that spiro[cyclopropane-1,3'-indolines] were formed by a multistep reaction process. According to the careful analysis of ^{1}H NMR data and comparison with the earlier reported results we could conclude that the two substituents at 2,3-position of cyclopropyl ring of spiro[cyclopropane-1,3'-indolines] existed in *trans*-configuration. This interesting result clearly indicated that the Michael-initiated cyclopropanation of pyridinium ylides is a diastereoselctive reaction.

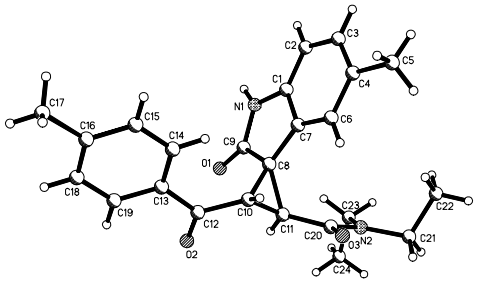
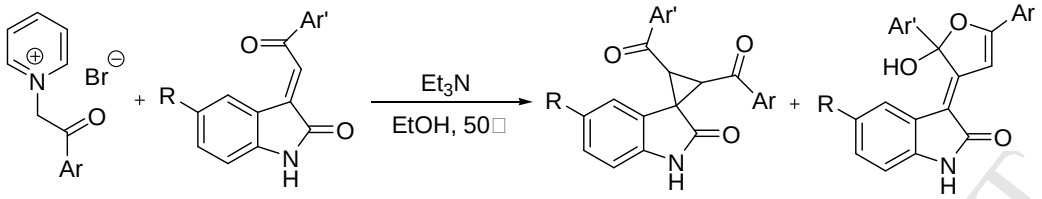


Figure 1 Molecular structure of spiro compound 2e

To explore the potential of this reaction as a practical protocol to obtain spiro[cyclopropane-1,3'-indolines], the triethylamine catalyzed reactions of *N*-phenacylpyridinium bromide (**1c**) with 3-phenacylideneoxindoles were also carried out in ethanol. The outcome of this reaction is a little surprise to us. Besides giving the normal spiro[cyclopropane-1,3'-indolines] (**4a-4g**) as main products, the functionalized 3-(2-hydroxyfuran-3(2H)-ylidene)oxindoles (**5a-5g**) were also produced as byproducts (Table 2, entries 1-7). In one case compound **5f** became the main product (72%). Trying to eliminate this byproduct by adjusting the reaction conditions such as solvent, base and temperature was not successful. The reactions usually resulted in a mixture of two products with little variable ratios. When *N*-*p*-chlorophenacylpyridinium bromide (**1d**) was employed in the reactions, a mixture of spiro compounds (**4h-4q**) and furanylideneoxindoles (**5h-5q**) were also obtained in different ratios (Table 2, entries 8-17). The structures of the prepared compounds were characterized with spectroscopic methods and confirmed by the X-ray diffraction determination of single crystals **4a** (Figure 2) and **5b** (Figure 3). ^{1}H NMR data of the prepared spiro[cyclopropane-1,3'-indolines] (**4a-4q**) also indicated that one diaseteroisomer predominately existed in most cases, while a mixture of *cis/trans*-isomers existed in samples **4j** and **4m**. Single crystal structure of compound **4a** clearly showed that two benzoyl groups are in *trans*-configuration.

Table 2 Reactions of *N*-phenacylpyridinium bromide with 3-phenacylideneoxindoles



				4		5	
Entry	Ar	R	Ar’	Compd	Yield (%)	Compd	Yield (%)
1	C_6H_5	H	C_6H_5	**4a**	54	**5a**	28
2	C_6H_5	H	*p*-$CH_3C_6H_4$	**4b**	48	**5b**	31
3	C_6H_5	CH_3	C_6H_5	**4c**	56	**5c**	30
4	C_6H_5	CH_3	*p*-$CH_3C_6H_4$	**4d**	55	**5d**	36
5	C_6H_5	F	C_6H_5	**4e**	58	**5e**	34
6	C_6H_5	Cl	C_6H_5	**4f**	—	**5f**	72
7	C_6H_5	Cl	*p*-$CH_3C_6H_4$	**4g**	62	**5g**	—
8	*p*-ClC_6H_4	H	C_6H_5	**4h**	70	**5h**	—
9	*p*-ClC_6H_4	H	*p*-ClC_6H_4	**4i**	42	**5i**	50
10	*p*-ClC_6H_4	H	*p*-$CH_3C_6H_4$	**4j**	72 (5:2)	**5j**	—
11	*p*-ClC_6H_4	CH_3	C_6H_5	**4k**	60	**5k**	26
12	*p*-ClC_6H_4	CH_3	*p*-ClC_6H_4	**4l**	43	**5l**	44
13	*p*-ClC_6H_4	CH_3	*p*-$CH_3C_6H_4$	**4m**	32 (4:1)	**5m**	47
14	*p*-ClC_6H_4	F	C_6H_5	**4n**	76	**5n**	—
15	*p*-ClC_6H_4	F	*p*-ClC_6H_4	**4o**	80	**5o**	—
16	*p*-ClC_6H_4	Cl	C_6H_5	**4p**	—	**5p**	78
17	*p*-ClC_6H_4	Cl	*p*-ClC_6H_4	**4q**	49	**5q**	32

a. isolate yields; b. pyridinium salts (1.1 mmol), 3-phenacylideneoxindole (1.0 mmol), triethylamine (0.2 mmol), EtOH (15 mL), 50°C, 6 h.

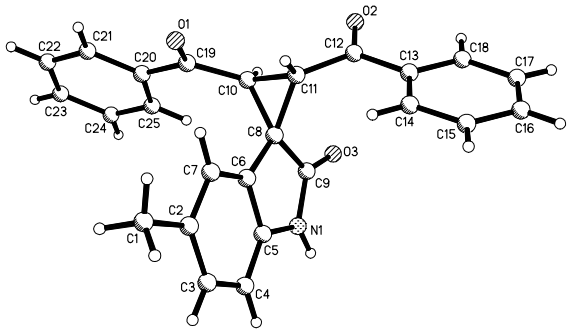


Figure 2 Molecular structure of compound **4a**

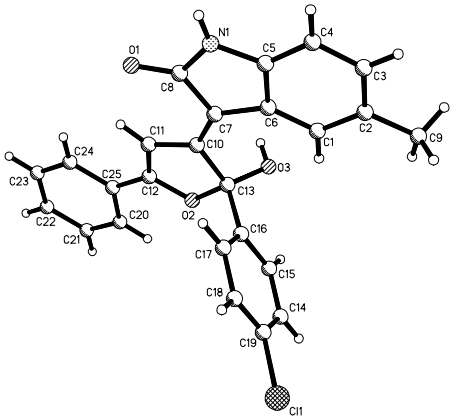
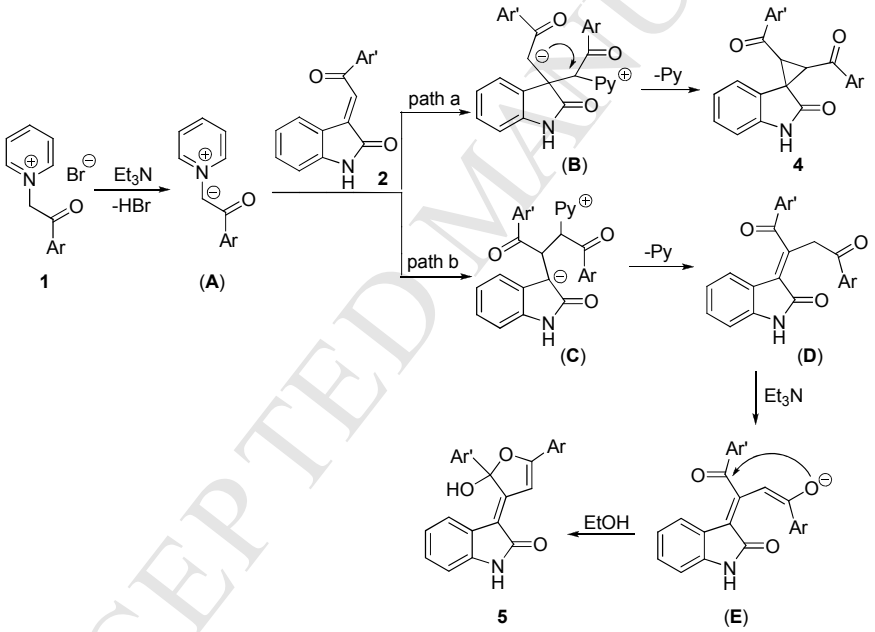


Figure 3 Molecular structure of compound **5b**

The formation of furanylideneoxindoles (**5a-5q**) in the reactions is unusual and very interesting. To explain the formation mechanism of the two products in the reactions, a plausible reaction mechanism was proposed in **Scheme 1**. At first the pyridinium salt is deprotonated by triethylamine to give a pyridinium ylide (**A**). Secondly Michael addition of pyridinium ylide (**A**) to 3-phenacylideneoxindole **2** undergoes in two different paths. In the path a, Michael addition of ylide (**A**) to the carbon atom at 3-position of 3-phenacylideneoxindole produces the intermediate (**B**). Then the intramolecular substitution of the carbanion to replace pyridine affords the final spiro[cyclopropane-1,3'-indolines] **4**. In the path b, Michael addition of ylide (**A**) to the exocyclic carbon atom of 3-phenacylideneoxindole gives another intermediate (**C**), which in turn transformed to a alkyleneoxindole intermediate (**E**) by sequential elimination of pyridine (**D**) and the coupling of the carbonium cation with carbanium ion to form C=C double bond. In the presence of triethylamine, intermediate (**E**) transferred to an enolate ion (**G**) by keto-enol tautomerism and deprotonation process. Finally the intramolecular nucleophilic addition of enolate ion to carbonyl group produces the furanylideneoxindole **5**. 3-phenacylideneoxindoles usually take part in the reactions at C-3 of oxindole moiety to give the versatile spirocyclic

oxindoles. it is unusual to observe the reactions taking place at the exocyclic carbon atom to give the isatinyl substituted products.[23, 24] It might be due to more steric hindrance at 3-position of oxindole moiety, the partially reaction of carbanium ion at exocyclic carbon atom resulted in the furanylideneoxindole **5** as byproduct. The similar reaction has been observed in the base catalyzed addition of malononitrile to 3-phencylideneoxindoles, in which the attack of carbanion of malononitrile at the *exo*-methylene atom of 3-phenacylideneoxindoles firstly gave a Michael adduct, then this Michael adduct transfered to the furanylideneoxindoles by cycloaddition of the enolized phenacyl group to one of cyano groups.[25]

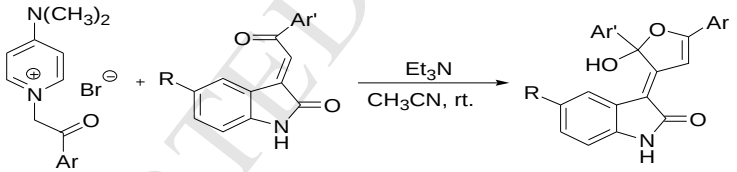


Scheme 1 The proposed reaction mechanism for the formation of compounds 4 and 5

In order to develop an efficient synthetic procedure for the functionalized 3-(2-hydroxyfuran-3(2H)-ylidene)oxindoles, many research works were carried out. We were pleased to find that under similar conditions the reactions of *N*-phenacyl- and *N*-*(*4-chlorophenacyl)-4-dimethylaminopyridinium bromides (**1e, 1f**) with 3-phenacylideneoxindole afforded predominately 3-(2-hydroxyfuran-3(2H)-ylidene)oxindoles in

very high yields (**Table 3**). All the reactions proceeded very smoothly and no spiro[cyclopropane-1,3'-indolines] were dictated in these reactions. The desired 3-(2-hydroxyfuran-3(2H)-ylidene)oxindoles were obtained in pure form after collection of the precipitates by filtration. In Table 3, five spiro compounds (**5c**, **5d**, **5k**, **5l**, **5m**) were the same products obtained in above reactions of pyridinium salt and the last three spiro compounds (**5r**, **5s**, **5t**) were prepared in this reaction. Their structures were established by the spectroscopic methods and the single crystal structure of compound **5l** (**Figure 4**) was also determined by X-ray diffraction. At present the reason for the formation of 3-(2-hydroxyfuran-3(2H)-ylidene)oxindole as sole product in the reaction of 4-dimethylaminopyridinium salts is very clear, but the facts that 4-dimethylaminopyridinium salts showed different reactivity to that of pyridinium salts in similar reaction are noticed in some reported works.[26]

Table 3 Reactions of *N*-phenacyl-4-dimethylaminopyridinium bromide with 3-phenacylideneoxindoles



5

Entry	Compd	Ar	R	Ar'	Yield (%)
1	**5c**	C_6H_5	CH_3	C_6H_5	88
2	**5d**	C_6H_5	CH_3	*p*-$CH_3C_6H_4$	80
3	**5k**	*p*-ClC_6H_4	CH_3	C_6H_5	87
4	**5l**	*p*-ClC_6H_4	CH_3	*p*-ClC_6H_4	83
5	**5m**	*p*-ClC_6H_4	CH_3	*p*-$CH_3C_6H_4$	85
6	**5r**	*p*-ClC_6H_4	CH_3	C_6H_5	80
7	**5s**	*p*-ClC_6H_4	Cl	*p*-$CH_3C_6H_4$	90
8	**5t**	*p*-ClC_6H_4	F	*p*-$CH_3C_6H_4$	92

a: isolate yields; b: 4-dimthylaminopyridinium salts (1.1 mmol), 3-phenacylidene oxindole (1.0 mmol), triethylamine (0.2 mmol), EtOH (15 mL), 50°C, 6 h.

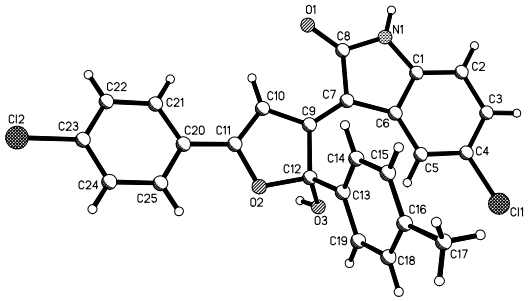


Figure 4 Molecular structure of compound **5l**

In conclusion we have systematically investigated the reactions of a series of pyridinium salts with various 3-phenacylidendoxindoles and found very interesting molecular diversity of Michael-initiated ring closure reactions of pyridinium ylides. Thus the efficient synthetic protocols for the functionalized spiro[cyclopropane-1,3'-indolines], and 3-furan-3(2H)-ylidene)indolin-2-ones were successfully developed. The scope and limitation of this reaction was established and the reaction mechanism was briefly discussed. These protocols have advantages of mild reaction conditions, easily accessible starting material and easy purification of the products, which makes it a useful and attractive method for the synthesis of the complex oxindole derivatives in synthetic and medicinal chemistry. Further expansion of the reaction scope and synthetic applications of this methodology are in progress in our laboratory.

Experimental Section

All reagents and solvents were commercially available with analytical grade and used as received. All evaporations of organic solvents were carried out with a rotary evaporator in conjunction with a water aspirator. Melting points were taken on a hot-plate microscope apparatus and were uncorrected. ^{1}H and ^{13}C NMR spectra were recorded with a Bruker AV-600 instrument. IR spectra were obtained on a Bruker Tensor 27 spectrometer (KBr disc). HRMS were measured at Bruker UHR-TOF maXis spectrometer. X-ray data were collected on a Bruker Smart APEX-2 diffractometer. Pyridinium salts were prepared by heating pyridine with *p*-nitrobenzyl bromide, *α*-phenacyl bromide and *N*,*N*-diethyl chloroacetamide in acetonitrile according to the published

procedure.[13]

1. General procedure for the preparation of spiro[cyclopropane-1,3'-indolines] 3a-3j from the reactions of pyrididinium salts with 3-phenacylideneoxindoles: A mixture of *N*-*p*-nitrobenzylpyridinium bromide or *N*-diethylcarbamoylmethylpyridinium chloride (1.1 mmol) 3-phenacylideneoxindoles (1.0 mmol) and triethylamine (0.2 mmol) in ethanol (15mL) was stirred at about 50°C for six hours. The resulting precipitates were collected by filtration, which were recrystallized in a mixture of chloroform and ethanol to give pure product for analysis.

2-benzoyl-3-(4-nitrophenyl)spiro[cyclopropane-1,3'-indolin]-2'-one (3a): White solid, 78%. m.p. 216-218□. IR (KBr): 3422, 3081, 1700, 1621, 1600, 1515, 1469, 1345, 1281, 1108, 1017, 975, 847. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.25 (s, 1H, NH), 8.18 (d, *J* = 8.4Hz, 2H, ArH) , 7.96 (d, *J* = 7.8Hz, 2H, ArH), 7.58 (t, *J* = 7.2Hz, 1H, ArH), 7.53 (d, *J* = 8.4Hz, 2H, ArH), 7.44 (t, *J* = 7.2Hz, 2H, ArH), 7.25 (d, *J* = 7.8Hz, 1H, ArH), 7.19 (t, *J* = 7.8Hz, 1H, ArH), 6.99 (t, *J* = 7.2Hz, 1H, ArH), 6.84 (d, *J* = 7.2Hz, 1H, ArH), 4.30 (d, *J* = 7.8Hz, 1H, CH), 4.17 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 192.3, 173.9, 147.2, 141.3, 141.2, 136.7, 134.0, 130.3, 128.9, 128.5, 128.1, 125.6, 123.3, 122.6, 122.3, 110.2, 42.0, 41.0, 38.3. HRMS (ESI) Calcd. for $C_{23}H_{16}N_2NaO_4$ ($[M+Na]^+$): 407.1002. Found: 407.1003.

2-(4-chlorobenzoyl)-3-(4-nitrophenyl)spiro[cyclopropane-1,3'-indolin]-2'-one (3b): White solid, 76%. m.p. 222-224□. IR (KBr): 3444, 1721, 1671, 1629, 1525, 1469, 1047, 1349, 1191, 1089, 1012, 842. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.19 (d, *J* = 9.0Hz, 2H, ArH), 7.90 (d, *J* = 9.0Hz, 2H, ArH), 7.81 (s, 1H, NH), 7.52 (d, *J* = 8.4Hz, 2H, ArH), 7.41 (d, *J* = 8.4Hz, 2H, ArH), 7.12 (t, *J* = 7.2Hz, 2H, ArH), 7.00 (t, *J* = 7.8Hz, 1H, ArH), 6.88 (d, *J* = 7.8Hz, 1H, ArH), 4.23 (d, *J* = 7.8Hz, 1H, CH), 4.15 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.0, 172.9, 147.3, 140.7, 134.8, 130.2, 129.8, 129.3, 128.3, 125.3, 123.4, 122.8, 122.3, 110.0, 41.7, 40.9, 38.4. HRMS (ESI) Calcd. for $C_{23}H_{15}ClN_2NaO_4$ ($[M+Na]^+$): 441.0613. Found: 441.0612.

2-benzoyl-5'-fluoro-3-(4-nitrophenyl)spiro[cyclopropane-1,3'-indolin]-2'-one (3c): White solid, 65%. m.p. 204-206□. IR (KBr): 3438, 3383, 1727, 1667, 1637, 1636, 1601, 1522, 1481, 1351, 1325, 1221, 1178, 1147, 1110. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.19 (d, *J* = 7.8Hz, 2H, ArH), 7.99 (d, *J* = 7.8Hz, 3H, ArH, NH), 7.61 (t, *J* = 7.2Hz, 1H, ArH), 7.53 (d, *J* = 8.4Hz, 2H, ArH), 7.48 (t, *J* = 7.2Hz, 2H, ArH), 7.08 (d, *J* = 6.6Hz, 1H, ArH), 6.91 (t, *J* = 8.4Hz, 1H, ArH), 6.79-6.77 (m, 1H, ArH), 4.33 (d, *J* = 7.8Hz, 1H, CH), 4.15 (d, *J* = 7.8Hz, 1H, CH) . ^{13}C NMR

(150MHz, $CDCl_3$) δ(ppm): 191.8, 173.0, 158.5 (d, *J* = 239.0Hz) , 147.4, 140.5, 136.7, 136.5, 134.2, 130.3, 129.0, 128.5, 127.1 (d, *J* = 9.0Hz), 122.4, 114.6 (d, *J* = 25.1Hz), 110.8 (d, *J* = 26.0Hz), 110.4 (d, *J* = 8.0Hz), 41.9, 40.9, 39.0. HRMS (ESI) Calcd. for $C_{23}H_{15}FN_2NaO_4$ ($[M+Na]^+$): 425.0908. Found: 425.0906.

2-(4-chlorobenzoyl)-5'-fluoro-3-(4-nitrophenyl)spiro[cyclopropane-1,3'-indolin]-2'-one (3d): White solid, 70%. m.p. 236-238□. IR (KBr): 3448, 1726, 1675, 1637, 1625, 1601, 1520, 1471, 1340, 1229, 1178, 1080, 1110, 857. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.27 (s, 1H, NH), 8.13 (d, *J* = 8.4Hz, 2H, ArH), 7.94 (d, *J* = 8.4Hz, 2H, ArH), 7.63 (d, *J* = 8.4Hz, 2H, ArH), 7.56 (d, *J* = 8.4Hz, 2H, ArH), 7.31 (dd, J_1= 2.4Hz, J_2 = 8.4Hz, 1H, ArH), 7.18 (dt, J_1= 1.8Hz, J_2 = 8.4Hz, 1H, ArH), 7.05 (dd, J_1= 3.6Hz, J_2 = 7.8Hz, 1H, ArH), 4.54 (d, *J* = 7.8Hz, 1H, ArH), 4.28 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.0, 173.5, 159.6 (d, *J* = 239.6Hz), 141.4, 141.0, 137.8, 135.2, 134.7, 130.5, 130.2, 129.8, 125.9 (d, *J* = 9.0Hz), 116.0 (d, *J* = 23.7Hz), 111.7 (d, *J* = 8.1Hz), 111.4 (d, *J* = 26.3Hz), 41.7, 40.3, 39.0. HRMS (ESI) Calcd. for $C_{23}H_{14}FClN_2NaO_4$ ($[M+Na]^+$): 483.1470. Found: 483.1474.

***N*,*N*-diethyl-5'-methyl-3-(4-methylbenzoyl)-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carb oxamide (3e):** White solid, 72%. m.p. 174-176□. IR (KBr): 3463, 2975, 1723, 1693, 1635, 1488, 1457, 1422, 1321, 1208, 1180, 1144, 1079, 1029, 985, 822. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.37 (s, 1H, NH), 7.72 (d, *J* = 7.8Hz, 2H, ArH), 7.14 (s, 2H, ArH), 7.13 (s, 1H, ArH), 7.05 (d, *J* = 7.8Hz, 1H, ArH), 6.78 (d, *J* = 7.8Hz, 1H, ArH), 3.97 (d, *J* = 7.8Hz, 1H, CH), 3.56-3.50 (m, 2H, 2CH), 3.24-3.15 (m, 3H, CH, CH_2), 2.33 (s, 3H, CH_3), 2.32 (s, 3H, CH_3), 1.03 (t, *J* = 7.2Hz, 3H, CH_3), 0.95 (t, *J* = 7.2Hz, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.6, 173.8, 164.5, 144.3, 139.0, 133.9, 132.0, 129.4, 128.7, 128.5, 125.0, 122.6, 110.0, 42.2, 40.8, 39.7, 38.7, 35.0, 21.6, 21.2, 14.0, 13.1. HRMS (ESI) Calcd. for $C_{24}H_{26}N_2NaO_3$ ($[M+Na]^+$): 413.2662. Found: 413.2658.

3-(4-chlorobenzoyl)-*N*,*N*-diethyl-5'-methyl-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carb oxamide (3f): White solid, 88%. m.p. 160-162□. IR (KBr):3443, 3233, 2972, 1720, 1628, 1591, 1488, 1424,1318, 1201, 1145, 1086, 1034, 911, 858. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.09 (s, 1H, NH), 7.76 (d, *J* = 7.8Hz, 2H, ArH), 7.33 (d, *J* = 7.8Hz, 2H, ArH), 7.15 (s, 1H, ArH), 7.07 (d, *J* = 7.8Hz, 1H, ArH), 6.80 (d, *J* = 7.8Hz, 1H, ArH), 3.95 (d, *J* = 7.8Hz, 1H, CH), 3.58-3.52 (m, 2H, 2CH), 3.24-3.16 (m, 3H, CH, CH_2), 2.32 (s, 3H, CH_3), 1.04 (t, *J* = 7.2Hz, 3H, CH_3), 0.96 (t, *J* =

7.2Hz, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 189.9, 173.6, 164.2, 139.9, 139.0, 134.6, 132.3, 129.8, 129.0, 128.9, 124.7, 122.7, 110.1, 42.2, 40.8, 39.5, 38.6, 35.0, 21.2, 14.0, 13.2. HRMS (ESI) Calcd. for $C_{23}H_{23}ClN_2NaO_3$ ($[M+Na]^+$): 433.1289. Found: 433.1290.

2-benzoyl-*N*,*N*-diethyl-5'-fluoro-2'-oxospiro[cyclopropane-1,3'-indoline]-3-carboxamide (3g): White solid, 69%. m.p. 200-202□. IR (KBr): 3366, 3276, 3068, 2972, 2934, 1708, 1637, 1484, 1423, 1322, 1223, 1156, 1089, 1026, 959, 832. ^1H NMR (600MHz, $CDCl_3$) δ(ppm): 8.85 (s, 1H, NH), 7.80 (d, *J* = 7.8Hz, 2H, ArH), 7.49 (t, *J* = 7.8Hz, 1H, ArH), 7.35 (t, *J* = 7.8Hz, 1H, ArH), 7.19 (dd, J_1 = 8.4Hz, J_2= 2.4Hz, 1H, ArH), 6.98 (td, J_1 = 8.4Hz, J_2= 2.4Hz, 1H, ArH) , 6.88 (dd, J_1 = 7.8Hz, J_2 = 1.8Hz, 1H, ArH), 4.03 (d, *J* = 7.8 Hz, 1H, CH), 3.58 (d, *J* = 7.8Hz, 1H, CH), 3.52-3.46 (m, 1H, CH), 3.32-3.27 (m, 1H, CH), 3.26-3.22 (m, 2H, CH_2) , 1.05 (t, *J* = 7.2Hz, 3H, CH_3), 0.97 (t, *J* = 7.2Hz, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.4, 173.8, 164.0, 159.8, 158.2, 137.5, 136.1, 133.6, 128.8, 128.4, 126.6, 126.5, 115.0, 114.8, 111.1, 111.0, 110.5, 110.3, 42.3, 40.9, 40.1, 38.9, 35.1, 14.2, 13.2. HRMS (ESI) Calcd. for $C_{22}H_{21}FN_2NaO_3$ ($[M+Na]^+$): 403.1428. Found: 403.1432.

3-(4-chlorobenzoyl)-*N*,*N*-diethyl-5'-fluoro-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carboxamide (3h): White solid, 54%. m.p. 230-232□. IR (KBr): 3451, 2970, 1731, 1634, 1592, 1485, 1419, 1223, 1183, 1148, 1091, 1016, 957, 822. ^1H NMR (600MHz, $CDCl_3$) δ(ppm): 8.34 (s, 1H, NH), 7.75 (d, *J* = 7.8Hz, 2H, ArH), 7.35 (d, *J* = 7.8Hz, 2H, ArH), 7.20 (d, *J* = 8.4Hz, 1H, ArH), 7.00 (t, *J* = 8.4Hz, 1H, ArH), 6.87 (dd, J_1 = 7.8Hz, J_2 = 3.6Hz, 1H, ArH), 3.99 (d, *J* = 7.8Hz, 1H, CH), 3.57 (d, *J* = 7.8Hz, 1H, CH), 3.52-3.47 (m, 1H, CH), 3.33-3.29 (m, 1H, CH), 3.27-3.24 (m, 2H, CH_2), 1.07 (t, *J* = 7.2Hz, 3H, CH_3), 1.00 (t, *J* = 7.2Hz, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 189.4, 173.5, 163.9, 159.1 (d, *J* = 239.6 Hz) , 140.1, 137.3, 134.4, 129.7, 129.1, 126.3 (d, *J* = 9.6Hz), 115.0 (d, *J* = 23.9Hz), 110.9 (d, *J* = 7.8Hz), 110.5 (d, *J* = 26.3Hz) , 40.3, 41.0, 39.9, 38.8, 35.1, 14.2, 13.2. HRMS (ESI) Calcd. for $C_{22}H_{20}ClFN_2NaO_3$ ($[M+Na]^+$): 437.1039. Found: 437.1041.

5'-chloro-3-(4-chlorobenzoyl)-*N*,*N*-diethyl-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carboxamide (3i): White solid, 82%. m.p. 242-244□. IR (KBr): 3420, 2982, 1703, 1631, 1591, 1480, 1420, 1322, 1270, 1208, 1141, 1092, 825. ^1H NMR (600MHz, $CDCl_3$) δ(ppm): 8.58 (s, 1H, NH), 7.74 (d, *J* = 8.4Hz, 2H, ArH), 7.39 (s, 1H, ArH), 7.35 (d, *J* = 8.4Hz, 2H, ArH), 7.26 (d, *J* = 8.4Hz, 1H, ArH), 6.88 (d, *J* = 8.4Hz, 1H, ArH), 4.00 (d, *J* = 7.8Hz, 1H, CH), 3.57-3.52 (m, 2H, 2CH),

3.28-3.19 (m, 3H, CH, CH_2), 1.06(t, *J* = 7.2Hz, 3H, CH_3), 0.99 (t, *J* = 7.2Hz, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 189.3, 173.3, 163.8, 130.2,139.9, 134.4,129.8, 129.2, 128.6, 128.3, 126.4, 122.7, 111.3, 42.3, 41.1, 39.8, 38.4, 35.1, 14.2, 13.2. HRMS (ESI) Calcd. for $C_{22}H_{20}Cl_2N_2NaO_3$ ($[M+Na]^+$): 453.0743. Found: 453.0745.

5'-chloro-*N*,*N*-diethyl-3-(4-methylbenzoyl)-2'-oxospiro[cyclopropane-1,3'-indoline]-2-car boxamide (3j): White solid, 80%. m.p. 166-168□. IR (KBr): 3460, 2981, 2026, 1703, 1633, 1420, 1322, 1271, 1208, 1141, 1085, 823. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.44 (s, 1H, NH), 7.69 (brs, 2H, ArH), 7.40 (s, 1H, ArH), 7.22 (brs, 1H, ArH), 7.14 (brs, 2H, ArH), 6.83 (brs, 1H, ArH), 4.02 (d, *J* = 6.6Hz, 1H, CH), 3.58 (d, *J* = 6.6Hz, 1H, CH), 3.53 (brs, 1H, CH), 3.22 (brs, 3H, CH, CH_2), 2.33 (s, 3H, CH_3), 1.05 (brs, 3H, CH_3), 0.96 (brs, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.1, 173.5, 163.9, 144.7, 139.8, 133.6, 129.5, 128.5, 128.4, 128.1, 126.7, 122.6, 111.2, 42.3, 41.0, 40.1, 38.5, 35.2, 21.7, 13.1, 13.2. HRMS (ESI) Calcd. for $C_{23}H_{23}ClN_2NaO_3$ ($[M+Na]^+$): 433.1289. Found: 433.1292.

2. General procedure for the preparation of spiro[cyclopropane-1,3'-indolines] 4a-4q and 3-furan-3(2H)-ylidene)indolin-2-ones 5a-5q from the reactions of pyrididinium salts with 3-phenacylideneoxindoles: A mixture of *N*-phenacylpyridinium bromide or *N*-*p*-chlorophenacylpyridinium chloride (1.1 mmol) 3-phenacylideneoxindoles (1.0 mmol) and triethylamine (0.2 mmol) in ethanol (15mL) was stirred at 50°C for six hours. The resulting precipitates were collected by filtration, which was bjected to thin-layer chromatography with a mixture of ethyl acetate and light petroleum (V/V = 2:1) to give the pure product for analysis.

(2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis(phenylmethanone) (4a): White solid, 54%. m.p. 176-178□. IR (KBr): 3256, 1719, 1673, 1620, 1595, 1465, 1400, 1308, 1219, 1085, 1020, 936, 801. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.53 (s, 1H, NH), 7.96 (d, *J* = 7.8Hz, 2H, ArH), 7.82 (d, *J* = 7.8Hz, 2H, ArH), 7.53 (t, *J* = 7.2Hz, 1H, ArH), 7.48 (t, *J* = 7.2Hz, 1H, ArH), 7.40 (t, *J* = 7.8Hz, 2H, ArH), 7.35-7.32 (m, 3H, ArH), 7.24 (t, *J* = 7.2Hz, 1H, ArH), 7.03 (t, *J* = 7.2Hz, 1H, ArH), 6.93 (d, *J* = 7.8Hz, 1H, ArH), 4.41 (d, *J* = 7.8Hz, 1H, CH), 4.17 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 192.1, 190.5, 173.6, 141.7, 136.7, 136.1, 133.8, 133.6, 128.8, 128.7, 128.6, 128.4, 124.3, 122.7, 122.5, 110.7, 41.3, 39.6, 38.7. HRMS (ESI) Calcd. for $C_{24}H_{17}NaNO_3$ ($[M+Na]^+$): 390.1101. Found: 390.1103.

3-(2-hydroxy-2,5-diphenylfuran-3(2H)-ylidene)indolin-2-one (5a): Yellow solid, 28%. m.p.

134-136□. IR (KBr): 3445, 3252, 1633, 1616, 1545, 1483, 1389, 1348, 1265, 1211, 1180, 1118, 969, 829. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.46 (s, 1H, NH), 8.91 (s, 1H, OH), 7.97 (s, 1H, ArH), 7.79 (d, *J* = 7.2Hz, 2H, ArH), 7.63 (d, *J* = 6.0Hz, 2H, ArH), 7.52-7.47 (m, 3H, ArH), 7.39-7.34 (m, 3H, ArH), 7.29 (s, 1H, ArH), 6.93 (t, *J* = 6.6Hz, 1H, ArH), 6.69 (d, *J* = 7.8Hz, 1H, ArH), 6.55 (t, *J* = 7.8Hz, 1H, ArH). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.8, 166.7, 155.3, 140.5, 138.1, 136.1, 133.3, 131.4, 129.0, 128.4, 126.5, 125.3, 122.8, 122.7, 120.0, 117.8, 111.9, 108.7, 100.6. HRMS (ESI) Calcd. for $C_{24}H_{17}NNaO_3$ ([M+Na]$^+$): 390.1101. Found: 390.1101.

2-benzoyl-3-(4-methylbenzoyl)spiro[cyclopropane-1,3'-indolin]-2'-one (4b): White solid, 48%. m.p. 192-194□. IR (KBr): 3281, 3021, 1726, 1670, 1601, 1470, 1406, 1306, 1218, 1094, 1019, 921, 832. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.52 (s, 1H, NH), 7.96 (d, *J* = 7.2Hz, 2H, ArH), 7.71 (d, *J* = 7.2Hz, 2H, ArH), 7.53 (t, *J* = 6.6Hz, 1H, ArH), 7.40 (t, *J* = 7.2Hz, 2H, ArH), 7.32 (d, *J* = 7.2Hz, 1H, ArH), 7.23 (t, *J* = 7.8Hz, 1H, ArH), 7.13 (t, *J* = 7.8Hz, 2H, ArH), 7.02 (t, *J* = 7.2Hz, 1H, ArH), 6.91 (d, *J* = 7.2Hz, 1H, ArH), 4.40 (d, *J* = 7.8Hz, 1H, CH), 4.15 (d, *J* = 7.8Hz, 1H, CH), 2.31 (s, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 192.2, 190.1, 173.6, 144.6, 141.6, 136.8, 133.8, 129.5, 128.8, 128.7, 128.6, 128.5, 128.4, 124.4, 122.7, 122.5, 110.6, 41.3, 39.7, 38.8, 21.6. HRMS (ESI) Calcd. for $C_{25}H_{19}NNaO_3$ ([M+Na]$^+$): 404.1257. Found: 404.1260.

3-(2-hydroxy-5-phenyl-2-p-tolylfuran-3(2H)-ylidene)indolin-2-one (5b): Yellow solid, 31%. m.p. 172-174□. IR (KBr): 3359, 1670, 1613, 1542, 1489, 1464, 1351, 1255, 1205, 1176, 1149, 1099, 1061, 957, 830. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.44 (s, 1H, NH), 8.83 (s, 1H, OH), 7.94 (s, 1H, ArH), 7.78 (d, *J* = 7.2Hz, 2H, ArH), 7.50-7.47 (m, 5H, ArH), 7.33 (d, *J* = 7.8Hz, 1H, ArH), 7.17 (d, *J* = 7.2Hz, 2H, ArH), 6.94 (t, *J* = 6.6Hz, 1H, ArH), 6.69 (d, *J* = 7.2Hz, 1H, ArH), 6.57 (t, *J* = 7.2Hz, 1H, ArH), 2.26 (s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.9, 166.6, 155.6, 140.5, 138.3, 135.3, 131.4, 129.0, 128.9, 128.7, 127.1, 127.0, 126.4, 125.3, 124.3, 121.8, 120.1, 114.8, 108.8, 100.6, 20.7. HRMS (ESI) Calcd. for $C_{25}H_{19}NNaO_3$ ([M+Na]$^+$): 404.1257. Found: 404.1258.

(5'-methyl-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis(phenylmethanone) (4c): White solid, 56%. m.p. 194-196□. IR (KBr): 3202, 1712, 1674, 1625, 1593, 1486, 1448, 1331, 1218, 1082, 1024, 941, 817. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.71 (s, 1H, NH), 7.95 (d, *J* = 7.2Hz, 2H, ArH), 7.80 (d, *J* = 7.2Hz, 2H, ArH), 7.52 (t, *J* = 7.2Hz, 1H, ArH), 7.46 (t, *J* = 7.8Hz, 1H, ArH), 7.39 (t, *J* = 7.8Hz, 2H, ArH), 7.32 (t, *J* = 7.8Hz, 2H, ArH), 7.11 (s, 1H, ArH), 7.03 (d, *J*

= 7.8Hz, 1H, ArH), 6.81 (d, *J* = 7.8Hz, 1H, ArH), 4.38 (d, *J* = 7.8Hz, 1H, CH), 4.12 (d, *J* = 7.8Hz, 1H, CH), 2.31 (s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 191.9, 191.0, 172.2, 140.2, 136.3, 135.9, 134.0, 133.6, 130.4, 129.1, 128.9, 128.8, 128.0, 127.9, 124.0, 122.6, 109.8, 40.4, 38.5, 38.4, 20.8. HRMS (ESI) Calcd. for $C_{25}H_{19}NNaO_3$ ([M+Na]$^+$): 404.1257. Found: 404.1260.

3-(2-hydroxy-2,5-diphenylfuran-3(2H)-ylidene)-5-methylindolin-2-one (5c): Yellow solid, 30%. m.p. 178-180□. IR (KBr): 3404, 3196, 1658, 1614, 1580, 1484, 1449, 1386, 1324, 1255, 1203, 1147, 1002, 935, 816. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.35 (s, 1H, NH), 8.88 (s, 1H, OH), 7.97 (s, 1H, ArH), 7.79 (s, 2H, ArH), 7.64 (s, 2H, ArH), 7.50 (s, 3H, ArH), 7.37 (s, 3H, ArH), 7.16 (s, 1H, ArH), 6.75 (s, 1H, ArH), 6.59 (s, 1H, ArH), 1.97 (s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 169.0, 166.4, 155.3, 138.3, 138.2, 131.3, 129.0, 128.9, 128.3, 128.2, 127.5, 126.4, 125.4, 125.1, 121.7, 115.1, 111.1, 108.4, 100.6, 20.8. HRMS (ESI) Calcd. for $C_{25}H_{19}NNaO_3$ ([M+Na]$^+$): 404.1257. Found: 404.1259.

2-benzoyl-5'-methyl-3-(4-methylbenzoyl)spiro[cyclopropane-1,3'-indolin]-2'-one (4d): White solid, 55%. m.p. 216-218□. IR (KBr): 3185, 3028, 1712, 1672, 1603, 1490, 1448, 1411, 1331, 1301, 1204, 1178, 1024, 808, 756. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.30 (s, 1H, NH), 7.96 (d, *J* = 7.8Hz, 2H, ArH), 7.71 (d, *J* = 8.4Hz, 2H, ArH), 7.53 (t, *J* = 7.2Hz, 1H, ArH), 7.40 (t, *J* = 7.8Hz, 2H, ArH), 7.14 (d, *J* = 7.8Hz, 2H, ArH), 7.11 (s, 1H, ArH), 7.02 (t, *J* = 7.8Hz, 1H, ArH), 6.78 (d, *J* = 7.8Hz, 1H, ArH), 4.37 (d, *J* = 7.8Hz, 1H, CH), 4.10 (d, *J* = 7.8Hz, 1H, CH), 2.32 (s, 6H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 192.3, 190.2, 173.4, 144.5, 139.1, 136.9, 133.9, 133.8, 132.3, 129.5, 129.0, 128.7, 128.6, 128.5, 124.5, 123.2, 110.1, 41.3, 39.8, 38.8, 21.6, 21.3. HRMS (ESI) Calcd. for $C_{26}H_{21}NNaO_3$ ([M+Na]$^+$): 418.1414. Found: 418.1416.

3-(2-hydroxy-5-phenyl-2-p-tolylfuran-3(2H)-ylidene)-5-methylindolin-2-one (5d): Yellow solid, 36%. m.p. 190-192□. IR (KBr): 3424, 3206, 1702, 1647, 1616, 1484, 1448, 1351, 1261, 1212, 1147, 1114, 1008, 936, 824. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.31 (s, 1H, NH), 8.80 (s, 1H, OH), 7.93 (s, 1H, ArH), 7.78 (d, *J* = 7.2Hz, 2H, ArH), 7.51-7.47 (m, 5H, ArH), 7.21 (s, 1H, ArH), 7.17 (d, *J* = 7.8Hz, 2H, ArH), 6.75 (d, *J* = 7.8Hz, 1H, ArH), 6.57 (d, *J* = 7.8Hz, 1H, ArH), 2.26 (s, 3H, CH_3), 1.99 (s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 169.0, 166.4, 155.5, 138.3, 135.4, 131.3, 129.0, 128.9, 128.8, 128.3, 127.5, 126.4, 125.3, 125.1,121.8, 114.9, 111.3, 108.4, 100.5, 20.8, 20.7. HRMS (ESI) Calcd. for $C_{26}H_{21}NNaO_3$ ([M+Na]$^+$): 418.1414. Found: 418.1414.

(5'-fluoro-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis(phenylmethanone) (4e): White solid, 58%. m.p. 188-190□. IR (KBr): 3313, 3019, 1715, 1673, 1598, 1485, 1331, 1222, 1184, 1151, 1017, 945, 739. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.84 (s, 1H, NH), 7.98 (d, *J* = 7.8Hz, 2H, ArH), 7.79 (d, *J* = 7.2Hz, 2H, ArH), 7.56 (t, *J* = 7.2Hz, 1H, ArH), 7.49 (t, *J* = 7.2Hz, 1H, ArH), 7.42 (d, *J* = 7.8Hz, 2H, ArH), 7.35 (t, *J* = 7.2Hz, 2H, ArH), 7.13 (d, *J* = 7.2Hz, 1H, ArH), 6.96 (t, *J* = 7.2Hz, 1H, ArH), 6.89 (d, *J* = 7.8Hz, 1H, ArH), 4.42 (d, *J* = 7.8Hz, 1H, CH), 4.14 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.9, 190.2, 173.8, 159.1 (d, *J* = 238.8Hz), 137.9, 136.5, 136.0, 134.1, 133.8, 128.9, 128.7, 128.4, 125.8 (d, *J* = 9.2Hz), 115.4 (d, *J* = 23.2Hz), 111.6 (d, *J* = 7.9Hz), 110.7 (d, *J* = 26.4Hz), 41.5, 39.9, 38.6. HRMS (ESI) Calcd. for $C_{24}H_{16}FNNaO_3$ ($[M+Na]^+$): 408.1006. Found: 408.1010.

5-fluoro-3-(2-hydroxy-2,5-diphenylfuran-3(2H)-ylidene)indolin-2-one (5e): Yellow solid, 34%. m.p. 170-172□. IR (KBr): 3404, 1717, 1671, 1604, 1537, 1479, 1388, 1347, 1287, 1252, 1216, 1180, 1148, 936, 817. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.49 (s, 1H, NH), 9.05 (s, 1H, OH), 7.95 (s, 1H, ArH), 7.82 (d, *J* = 7.2Hz, 2H, ArH), 7.63 (d, *J* = 6.6Hz, 2H, ArH), 7.54 (d, *J* = 6.6Hz, 1H, ArH), 7.51 (d, *J* = 7.2Hz, 2H, ArH), 7.40 (d, *J* = 7.2Hz, 3H, ArH), 7.06 (d, *J* = 9.0Hz, 1H, ArH), 6.78 (t, *J* = 7.2Hz, 1H, ArH), 6.67 (d, *J* = 7.2Hz, 1H, ArH). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.8, 167.8, 156.8 (d, *J* = 231.3Hz), 157.0, 137.7, 136.7, 131.8, 129.3, 129.1, 128.6, 126.7, 125.2, 122.7 (d, *J* = 9.3Hz), 114.2, 113.2 (d, *J* = 23.4Hz), 111.3, 110.7 (d, *J* = 26.9Hz), 109.2 (d, *J* = 8.9Hz), 100.6. HRMS (ESI) Calcd. for $C_{24}H_{16}FNNaO_3$ ($[M+Na]^+$): 408.1006. Found: 408.1006.

5-chloro-3-(2-hydroxy-2,5-diphenylfuran-3(2H)-ylidene)indolin-2-one (5f): Yellow solid, 72%. m.p. 156-158□. IR (KBr): 3449, 3404, 1657, 1539, 1480, 1417, 1386, 1349, 1320, 1249, 1201, 1144, 933, 814. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.60 (s, 1H, NH), 9.07 (s, 1H, OH), 7.94 (s, 1H, ArH), 7.82 (d, *J* = 7.2Hz, 2H, ArH), 7.62 (d, *J* = 6.6Hz, 2H, ArH), 7.54 (d, *J* = 6.6Hz, 1H, ArH), 7.50 (t, *J* = 7.2Hz, 2H, ArH), 7.39 (d, *J* = 6.6Hz, 3H, ArH), 7.31 (s, 1H, ArH), 6.98 (d, *J* = 7.8Hz, 1H, ArH), 6.70 (d, *J* = 7.8Hz, 1H, ArH). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.6, 168.1, 157.4, 139.0, 137.7, 131.8, 129.3, 129.1, 128.6, 128.5, 126.7, 126.4, 125.3, 124.2, 123.6, 123.3, 113.6, 111.4, 110.0, 100.7. HRMS (ESI) Calcd. for $C_{24}H_{16}ClNNaO_3$ ($[M+Na]^+$): 424.0711. Found: 424.0710.

2-benzoyl-5'-chloro-3-(4-methylbenzoyl)spiro[cyclopropane-1,3'-indolin]-2'-one (4g):

White solid, 62%. m.p. 222-224□. IR (KBr): 3179, 3063, 3022, 2862, 1723, 1667, 1606, 1477, 1445, 1327, 1221, 1088, 1023, 937, 809. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.56 (s, 1H, NH), 7.99 (d, *J* = 7.8Hz, 2H, ArH), 7.69 (d, *J* = 7.8Hz, 2H, ArH), 7.56 (t, *J* = 7.2Hz, 1H, ArH), 7.43 (t, *J* = 7.2Hz, 2H, ArH), 7.36 (t, *J* = 6.6Hz, 1H, ArH), 7.22 (d, *J* = 7.8Hz, 1H, ArH), 7.15 (d, *J* = 7.8Hz, 2H, ArH), 6.86 (d, *J* = 8.4Hz, 1H, ArH), 4.40 (d, *J* = 7.8Hz, 1H, CH), 4.12 (d, *J* = 7.8Hz, 1H, CH), 2.33 (s, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.8, 189.7, 173.4, 144.9, 140.2, 136.6, 134.0, 133.5, 129.6, 129.5, 128.8, 128.8, 128.5, 128.4, 128.2, 126.1, 123.1, 111.7, 41.1, 40.1, 38.8, 21.7. HRMS (ESI) Calcd. for $C_{25}H_{18}ClNNaO_3$ ([M+Na]$^+$): 438.0867. Found: 438.0870.

2-benzoyl-3-(4-chlorobenzoyl)spiro[cyclopropane-1,3'-indolin]-2'-one (4h): White solid, 70%. m.p. 208-210□. IR (KBr): 3283, 1725, 1670, 1621, 1598, 1469, 1398, 1318, 1221, 1092, 1014, 934, 842, 740. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.65 (s, 1H, NH), 7.98 (d, *J* = 7.8Hz, 2H, ArH), 7.81 (d, *J* = 7.8Hz, 2H, ArH), 7.49 (t, *J* = 7.2Hz, 1H, ArH), 7.36-7.34 (m, 4H, ArH), 7.29 (d, *J* = 7.8Hz, 1H, ArH), 7.25 (d, *J* = 7.2Hz, 1H, ArH), 7.03 (t, *J* = 7.8Hz, 1H, ArH), 6.94 (d, *J* = 7.8Hz, 1H, ArH), 4.35 (d, *J* = 8.4Hz, 1H, CH), 4.16 (d, *J* = 8.4Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.0, 190.4, 173.7, 141.8, 140.5, 136.0, 134.9, 133.8, 133.0, 129.1, 128.9, 128.8, 128.4, 124.1, 122.8, 122.4, 110.0, 41.3, 39.5, 38.5. HRMS (ESI) Calcd. for $C_{24}H_{16}ClNNaO_3$ ([M+Na]$^+$): 424.0711. Found: 424.0714.

(2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis((4-chlorophenyl)methanone) (4i): White solid, 42%. m.p. 202-204□. IR (KBr): 3292, 3053, 1711, 1672, 1618, 1586, 1470, 1397, 1315, 1220, 1191, 1092, 1010, 843. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.66 (s, 1H, NH), 7.88 (d, *J* = 8.4Hz, 2H, ArH), 7.73 (d, *J* = 8.4Hz, 2H, ArH), 7.35 (d, *J* = 8.4Hz, 2H, ArH), 7.33 (d, *J* = 8.4Hz, 2H, ArH), 7.28-7.25 (m, 2H, ArH), 7.03 (t, *J* = 7.8Hz, 1H, ArH), 6.95 (d, *J* = 7.2Hz, 1H, ArH), 4.32 (d, *J* = 7.2Hz, 1H, CH), 4.11 (d, *J* = 7.2Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.8, 189.4, 173.5, 141.7, 140.6, 140.3, 134.8, 134.4, 129.9, 129.8, 129.2, 129.1, 129.0, 123.9, 122.9, 122.5, 110.9, 41.2, 39.3, 38.5. HRMS (ESI) Calcd. for $C_{24}H_{15}Cl_2NNaO_3$ ([M+Na]$^+$): 458.0321. Found: 458.0324.

3-(2,5-bis(4-chlorophenyl)-2-hydroxyfuran-3(2H)-ylidene)indolin-2-one (5i): Yellow solid, 50%. m.p. 150-152□. IR (KBr): 3438, 3168, 1658, 1611, 1571, 1542, 1485, 1406, 1352, 1266, 1177, 1152, 1096, 1053, 928, 838. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.50 (s, 1H, NH), 9.07 (s, 1H, OH), 7.96 (s, 1H, ArH), 7.81 (d, *J* = 8.4Hz, 2H, ArH), 7.62 (d, *J* = 8.4Hz, 2H, ArH),

7.55 (d, *J* = 8.4Hz, 2H, ArH), 7.44 (d, *J* = 8.4Hz, 2H, ArH), 7.27 (d, *J* = 7.2Hz, 1H, ArH), 6.98 (t, *J* = 7.2Hz, 1H, ArH), 6.71 (d, *J* = 7.8Hz, 1H, ArH), 6.60 (t, *J* = 7.8Hz, 1H, ArH). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.7, 165.0, 154.2, 140.7, 137.0, 136.0, 133.7, 129.2, 128.6, 128.2, 127.6, 127.3, 124.2, 121.4, 120.2, 115.6, 115.5, 110.8, 109.0, 101.2. HRMS (ESI) Calcd. for $C_{24}H_{15}Cl_2NNaO_3$ ([M+Na]$^+$): 458.0321. Found: 458.0320.

2-(4-chlorobenzoyl)-3-(4-methylbenzoyl)2-(4-chlorobenzoyl)-3-(4-methyl-benzoyl)spiro[cyclopropane-1,3'-indolin]-2'-one (4j): White solid, 72%. m.p. 198-200□. IR (KBr): 3293, 3051, 1734, 1708, 1668, 1609, 1469, 1365, 1320, 1221, 1177, 1093, 1034, 1012, 841, 749. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): *E*-isomer: 8.62 (s, 1H, NH), 7.88 (d, *J* = 7.8Hz, 2H, ArH), 7.70 (d, *J* = 7.8Hz, 2H, ArH), 7.34 (d, *J* = 8.4Hz, 2H, ArH), 7.29 (d, *J* = 7.2Hz, 1H, ArH), 7.24 (d, *J* = 7.8Hz, 1H, ArH), 7.14 (d, *J* = 7.8Hz, 2H, ArH), 7.03 (t, *J* = 7.8Hz, 1H, ArH), 6.94 (d, *J* = 7.8Hz, 1H, ArH), 4.33 (d, *J* = 7.8Hz, 1H, CH), 4.14 (d, *J* = 7.8Hz, 1H, CH), 2.32 (s, 3H, CH_3); *Z*-isomer: 8.56 (s, 1H, NH), 7.85 (d, *J* = 7.8Hz, 2H, ArH), 7.75 (d, *J* = 7.8Hz, 2H, ArH), 7.32 (d, *J* = 8.4Hz, 2H, ArH), 7.17 (d, *J* = 7.8Hz, 1H, ArH), 4.36 (d, *J* = 7.8Hz, 1H, CH), 4.12 (d, *J* = 7.8Hz, 1H, CH), 2.35 (s, 3H, CH_3); *Z*/*E* = 2:5; ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.3, 191.0, 189.9, 189.6, 173.5, 145.0, 144.5, 141.6, 140.4, 140.2, 135.0, 134.6, 133.7, 129.9, 129.8, 129.5, 129.1, 128.7, 128.5, 124.2, 122.8, 122.5, 110.7, 41.3, 41.0, 39.7, 39.3, 38.7, 38.2, 31.7. HRMS (ESI) Calcd. for $C_{25}H_{18}ClNNaO_3$ ([M+Na]$^+$): 438.0867. Found: 438.0870.

2-benzoyl-3-(4-chlorobenzoyl)-5'-methylspiro[cyclopropane-1,3'-indolin]-2'-one (4k): White solid, 60%. m.p. 220-222□. IR (KBr): 3187, 3032, 1714, 1670, 1626, 1592, 1489, 1447, 1329, 1301, 1205, 1092, 1020, 807. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.30 (s, 1H, NH), 7.96 (d, *J* = 7.8Hz, 2H, ArH), 7.74 (d, *J* = 8.4Hz, 2H, ArH), 7.54 (t, *J* = 7.2Hz, 1H, ArH), 7.40 (t, *J* = 7.2Hz, 2H, ArH), 7.32 (d, *J* = 8.4Hz, 2H, ArH), 7.10 (s, 1H, ArH), 7.04 (t, *J* = 7.2Hz, 1H, ArH), 6.80 (d, *J* = 7.8Hz, 1H, ArH), 4.36 (d, *J* = 7.8Hz, 1H, CH), 4.07 (d, *J* = 7.8Hz, 1H, CH), 2.31 (s, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 192.0, 189.6, 173.1, 140.2, 139.0, 136.7, 134.5, 134.0, 132.5, 129.8, 129.2, 128.8, 128.6, 124.1, 123.2, 110.1, 41.2, 39.4, 38.7, 21.3. HRMS (ESI) Calcd. for $C_{25}H_{18}ClNNaO_3$ ([M+Na]$^+$): 438.0867. Found: 438.0869.

3-(2-(4-chlorophenyl)-2-hydroxy-5-phenylfuran-3(2H)-ylidene)-5-methylindol-in-2-one (5k): Yellow solid, 26%. m.p. 196-198□. IR (KBr): 3444, 3163, 1655, 1544, 1484, 1406, 1337, 1268, 1209, 1088, 1006, 936, 811. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.34 (s, 1H, NH),

8.89 (s, 1H, OH), 7.96 (s, 1H, ArH), 7.81 (d, *J* = 8.4Hz, 2H, ArH), 7.63 (d, *J* = 6.6Hz, 2H, ArH), 7.54 (d, *J* = 8.4Hz, 2H, ArH), 7.37 (d, *J* = 7.8Hz, 3H, ArH), 7.13 (s, 1H, ArH), 6.76 (d, *J* = 7.8Hz, 1H, ArH), 6.57 (d, *J* = 7.8Hz, 1H, ArH), 1.96(s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.9, 165.0, 154.8, 138.4, 138.1, 135.8, 129.1, 129.0, 128.4, 128.3, 128.2, 127.8, 127.7, 125.4, 125.1, 121.6, 115.6, 111.3, 108.5, 101.1, 20.7. HRMS (ESI) Calcd. for $C_{25}H_{18}ClNNaO_3$ ($[M+Na]^+$): 438.0867. Found: 438.0866.

(5'-methyl-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis((4-chlorophenyl)methanone) (4l): White solid, 43%. m.p. 182-184□. IR (KBr): 3442, 1709, 1670, 1590, 1490, 1403, 1326, 1202, 1092, 1042, 1012, 822, 763. ^1H NMR (600MHz, $CDCl_3$) δ(ppm): 8.25 (s, 1H, NH), 7.88 (d, *J* = 8.4Hz, 2H, ArH), 7.74 (d, *J* = 8.4Hz, 2H, ArH), 7.35-7.33 (m, 4H, ArH), 7.06 (s, 1H, ArH), 7.05 (d, *J* = 7.8Hz, 1H, ArH), 6.81 (d, *J* = 7.8Hz, 1H, ArH), 4.30 (d, *J* = 7.8Hz, 1H, CH), 4.07 (d, *J* = 7.8Hz, 1H, CH), 2.32 (s, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 192.8, 189.4, 173.2, 140.5, 140.3, 139.1, 134.9, 134.4, 132.6, 129.9, 129.8, 129.4, 129.2, 129.1, 123.9, 123.1, 110.3, 41.2, 39.3, 38.5, 21.3. HRMS (ESI) Calcd. for $C_{25}H_{17}Cl_2NNaO_3$ ($[M+Na]^+$): 472.0478. Found: 472.0478.

3-(2,5-bis(4-chlorophenyl)-2-hydroxyfuran-3(2H)-ylidene)-5-methylindolin-2-one (5l): Yellow solid, 44%. m.p. 152-154□. IR (KBr): 3435, 3199, 1653, 1545, 1485, 1405, 1338, 1266, 1211, 1149, 1095, 1052, 1009, 938, 842. ^1H NMR (600MHz, DMSO-d_6) δ(ppm): 10.37 (s, 1H, NH), 9.04 (s, 1H, OH), 7.95 (s, 1H, ArH), 7.81 (d, *J* = 8.4Hz, 2H, ArH), 7.63 (d, *J* = 7.8Hz, 2H, ArH), 7.54 (d, *J* = 8.4Hz, 2H, ArH), 7.45 (d, *J* = 7.8Hz, 2H, ArH), 7.13 (s, 1H, ArH), 6.79 (d, *J* = 7.8Hz, 1H, ArH), 6.69 (d, *J* = 7.8Hz, 1H, ArH), 1.99 (s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.8, 164.9, 154.1, 138.5, 137.1, 135.9, 133.7, 129.2, 128.5, 128.4, 128.2, 128.0, 127.6, 127.3, 125.0, 121.4, 115.8, 110.6, 108.6, 101.1, 20.8. HRMS (ESI) Calcd. for $C_{25}H_{17}Cl_2NNaO_3$ ($[M+Na]^+$): 472.0478. Found: 472.0474.

2-(4-chlorobenzoyl)-5'-methyl-3-(4-methylbenzoyl)spiro[cyclopropane-1,3'-indolin]-2'-one (4m): White solid, 32%. m.p. 212-214□. IR (KBr): 3297, 3023, 2917, 1717, 1674, 1597, 1489, 1400, 1325, 1184, 1093, 1039, 822. ^1H NMR (600MHz, $CDCl_3$) δ(ppm): *E*-isomer: 8.20 (s, 1H, NH), 7.89 (d, *J* = 8.4Hz, 2H, ArH), 7.70 (d, *J* = 7.8Hz, 2H, ArH), 7.34 (d, *J* = 8.4Hz, 2H, ArH), 7.16 (d, *J* = 7.8Hz, 2H, ArH), 7.08 (s, 1H, ArH), 7.04 (d, *J* = 7.8Hz, 1H, ArH), 6.80 (d, *J* = 7.2Hz, 1H, ArH), 4.31 (d, *J* = 7.8Hz, 1H, CH), 4.09 (d, *J* = 7.8Hz, 1H, CH), 2.34 (s, 3H, CH_3), 2.32 (s,

3H, CH_3). *Z*-isomer: 8.16 (s, 1H, NH), 7.85 (d, *J* = 7.8Hz, 2H, ArH), 7.75 (d, *J* = 7.8Hz, 2H, ArH), 7.18 (d, *J* = 8.4Hz, 2H, ArH), 4.34 (d, *J* = 7.8Hz, 1H, CH), 4.07 (d, *J* = 7.8Hz, 1H, CH), 2.36 (s, 3H, CH_3), 2.31 (s, 3H, CH_3); *Z/E* = 1:4. ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.5, 191.1, 190.2, 189.8, 173.7, 144.9, 144.7, 140.3, 140.1, 139.5, 135.0, 134.6, 134.2, 133.7, 132.3, 129.9, 129.8, 129.5, 129.3, 129.1, 128.7, 128.6, 124.2, 122.9, 110.7, 41.4, 41.2, 39.5, 39.2, 38.7, 38.6, 21.7, 21.3. HRMS (ESI) Calcd. for $C_{26}H_{20}ClNNaO_3$ ($[M+Na]^+$): 452.1024. Found: 452.1025.

3-(5-(4-chlorophenyl)-2-hydroxy-2-p-tolylfuran-3(2H)-ylidene)-5-methylindolin-2-one (5m): Yellow solid, 47%. m.p. 188-190□. IR (KBr): 3440, 3220, 1654, 1571, 1543, 1482, 1406, 1383, 1334, 1266, 1220, 1150, 1111, 1090, 935, 811. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.33 (s, 1H, NH), 8.83 (s, 1H, OH), 7.94 (s, 1H, ArH), 7.80 (d, *J* = 8.4Hz, 2H, ArH), 7.54 (d, *J* = 7.8Hz, 2H, ArH), 7.50 (d, *J* = 7.8Hz, 2H, ArH), 7.19 (d, *J* = 8.4Hz, 2H, ArH), 7.16 (s, 1H, ArH), 6.76 (d, *J* = 7.2Hz, 1H, ArH), 6.57 (d, *J* = 7.8Hz, 1H, ArH), 2.26 (s, 3H, CH_3), 1.98 (s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.9, 165.0, 155.0, 138.3, 135.7, 135.3, 129.1, 128.8, 128.4, 128.1, 127.9, 127.8, 127.7, 125.3, 121.7, 115.5, 111.4, 108.4, 105.4, 101.0, 20.8, 20.7. HRMS (ESI) Calcd. for $C_{26}H_{20}ClNNaO_3$ ($[M+Na]^+$): 452.1024. Found: 452.1022.

2-benzoyl-3-(4-chlorobenzoyl)-5'-fluorospiro[cyclopropane-1,3'-indolin]-2'-one (4n): White solid, 76%. m.p. 204-206□. IR (KBr): 3444, 3013, 1725, 1696, 1665, 1589, 1484, 1401, 1329, 1229, 1183, 1150, 1091, 1036, 949, 850. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.63 (s, 1H, NH), 7.92 (d, *J* = 8.4Hz, 2H, ArH), 7.79 (d, *J* = 7.8Hz, 2H, ArH), 7.51 (t, *J* = 7.2Hz, 1H, ArH), 7.40 (t, *J* = 8.4Hz, 2H, ArH), 7.37 (d, *J* = 7.2Hz, 2H, ArH), 7.11 (d, *J* = 7.8Hz, 1H, ArH), 6.98 (t, *J* = 8.4Hz, 1H, ArH), 6.89 (d, *J* = 7.8Hz, 1H, ArH), 4.36 (d, *J* = 7.8Hz, 1H, CH), 4.12 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.7, 190.0, 173.4, 159.1 (d, *J* = 239.3Hz), 140.7, 137.6, 135.9, 134.8, 133.9, 130.0, 129.2, 128.9, 12.84, 125.6 (d, *J* = 7.1Hz), 115.5 (d, *J* = 24.3Hz), 111.4 (d, *J* = 8.1Hz), 110.8 (d, *J* = 25.3Hz), 41.4, 40.0, 38.5. HRMS (ESI) Calcd. for $C_{24}H_{15}FClNNaO_3$ ($[M+Na]^+$): 442.0617. Found: 442.0619.

(5'-fluoro-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis((4-chlorophenyl)methanone) (4o): White solid, 80%. m.p. 200-202□. IR (KBr): 3251, 1721, 1677, 1587, 1480, 1400, 1324, 1224, 1179, 1092, 1011, 841, 811. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.07 (s, 1H, NH), 7.93 (d, *J* = 8.4Hz, 2H, ArH), 7.74 (d, *J* = 8.4Hz, 2H, ArH), 7.42 (t, *J* = 8.4Hz, 2H, ArH), 7.37 (t, *J* = 8.4Hz, 2H, ArH), 7.10 (d, *J* = 8.4Hz, 1H, ArH), 6.98 (t, *J* = 8.4Hz, 1H, ArH), 6.86 (d, *J* = 7.8Hz,

1H, ArH), 4.35 (d, *J* = 7.8Hz, 1H, CH), 4.08 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.5, 188.9, 173.0, 159.1 (d, *J* = 239.7Hz), 140.9, 140.5, 137.4, 134.7, 134.2, 130.0, 129.7, 129.5, 129.3, 125.4 (d, *J* = 7.5Hz), 115.5 (d, *J* = 23.7Hz), 111.2 (d, *J* = 8.1Hz), 110.9 (d, *J* = 26.3Hz), 41.2, 39.8, 38.5. HRMS (ESI) Calcd. for $C_{24}H_{14}FCl_2NNaO_3$ ($[M+Na]^+$): 476.0227. Found: 476.0229.

5-chloro-3-(5-(4-chlorophenyl)-2-hydroxy-2-phenylfuran-3(2H)-ylidene)indolin-2-one (5p): Yellow solid, 78%. m.p. 180-182□. IR (KBr): 3440, 3178, 1660, 1591, 1542, 1489, 1449, 1408, 1340, 1239, 1170, 1112, 1081, 981, 813. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.63 (s, 1H, NH), 9.11 (s, 1H, OH), 7.96 (s, 1H, ArH), 7.85 (d, *J* = 7.2Hz, 2H, ArH), 7.62 (d, *J* = 7.2Hz, 2H, ArH), 7.56 (d, *J* = 7.2Hz, 2H, ArH), 7.39 (brs, 3H, ArH), 7.30 (s, 1H, ArH), 6.99 (d, *J* = 7.2Hz, 1H, ArH), 6.70 (d, *J* = 7.2Hz, 1H, ArH). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.5, 166.6, 156.9, 139.1, 137.6, 136.3, 129.3, 129.2, 128.5, 128.4, 127.4, 126.5, 125.3, 124.2, 123.7, 123.1, 114.1, 111.5, 110.1, 101.2. $C_{24}H_{15}Cl_2NNaO_3$ ($[M+Na]^+$): 458.0321. Found: 458.0322.

(5'-chloro-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis((4-chlorophenyl)methanone) (4q): White solid, 49%. m.p. 210-212□. IR (KBr): 3290, 3049, 1721, 1672, 1618, 1594, 1472, 1399, 1315, 1220, 1191, 1092, 1020, 843. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.34 (s, 1H, NH), 7.92 (d, *J* = 8.4Hz, 2H, ArH), 7.73 (d, *J* = 8.4Hz, 2H, ArH), 7.42 (d, *J* = 8.4Hz, 2H, ArH), 7.36 (d, *J* = 8.4Hz, 2H, ArH), 7.32 (d, *J* = 7.8Hz, 1H, ArH), 7.25 (d, *J* = 7.8Hz, 1H, ArH), 6.88 (d, *J* = 7.8Hz, 1H, ArH), 4.33 (d, *J* = 7.8Hz, 1H, CH), 4.09 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.5, 188.8, 172.9, 140.9, 140.6, 140.0, 134.7, 134.2, 130.1, 129.7, 129.3, 129.2, 129.0, 128.5, 125.6, 123.1, 111.7, 40.9, 39.8, 38.5. HRMS (ESI) Calcd. for $C_{24}H_{14}Cl_3NNaO_3$ ($[M+Na]^+$): 491.9931. Found: 491.9932.

3-(2,5-bis(4-chlorophenyl)-2-hydroxyfuran-3(2H)-ylidene)-5-chloroindolin-2-one (5q): Yellow solid, 32%. m.p. 184-186□. IR (KBr): 3399, 3194, 1659, 1611, 1537, 1481, 1446, 1348, 1250, 1203, 1169, 1110, 1075, 975, 815. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.64 (s, 1H, NH), 9.24 (s, 1H, OH), 7.95 (s, 1H, ArH), 7.84 (d, *J* = 7.8Hz, 2H, ArH), 7.62 (d, *J* = 7.8Hz, 2H, ArH), 7.56 (d, *J* = 8.4Hz, 2H, ArH), 7.47 (d, *J* = 8.4Hz, 2H, ArH), 7.27 (s, 1H, ArH), 7.00 (s, 1H, ArH), 6.72 (d, *J* = 7.8Hz, 1H, ArH). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.7, 164.7, 154.0, 138.4, 137.0, 135.7, 133.5, 129.0, 128.4, 128.3, 128.0, 127.8, 127.5, 127.2, 124.8, 121.3, 115.6, 110.4, 108.5, 101.0. HRMS (ESI) Calcd. for $C_{24}H_{14}Cl_3NNaO_3$ ($[M+Na]^+$): 491.9931. Found:

491.9926.

3. General procedure for the preparation of 3-furan-3(2H)-ylidene)indolin-2-ones from the reactions of *N*-phenacyl-4-dimethylaminopyrididinium salts with 3-phenacylideneoxindoles: A mixture of *N*-phenacyl-4-dimethylaminopyridinium bromide or *N*-*p*-chlorophenacyl-4-dimethylaminopyridinium chloride (1.1 mmol) 3-phenacylideneoxindoles (1.0 mmol) and triethylamine (0.2 mmol) in acetonitrile (10mL) was stirred at room temperature for six hours. The resulting precipitates were collected by filtration, which was collected by filtration and washed with cold ethanol to give pure product for analysis.

3-(5-(4-chlorophenyl)-2-hydroxy-2-phenylfuran-3(2H)-ylidene)-5-methylindolin-2-one (5r): Red solid, 80%. m.p. 216-218□. IR (KBr): 3433, 2025, 1671, 1569, 1545, 1481, 1454, 1427, 1405, 1383, 1346, 1322, 1257, 1196, 1151, 1085, 1067, 1007, 933, 880, 821, 802, 767, 733, 699, 644. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.36 (s, 1H, NH), 9.01 (s, 1H, OH), 7.99 (s, 1H, CH), 7.81 (d, *J* = 8.4Hz, 2H, ArH), 7.65 (d, *J* = 7.2Hz, 2H, ArH), 7.54 (d, *J* = 8.4Hz, 2H, ArH), 7.38 (q, *J* = 7.8Hz, 3H, ArH), 7.14 (s, 1H, ArH), 6.76 (d, *J* = 7.8Hz, 1H, ArH), 6.58 (d, *J* = 7.8Hz, 1H, ArH), 1.97 (s, 3H, CH3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.9, 165.2, 154.8, 138.3, 138.1, 135.8, 129.1, 129.0, 128.4, 128.3, 128.2, 127.6, 125.4, 125.0, 121.7, 115.3, 108.4, 101.0,20.7. HRMS (ESI) Calcd. for $C_{25}H_{18}ClNNaO_3$ ($[M+Na]^+$): 438.0867. Found: 438.0865.

5-chloro-3-(5-(4-chlorophenyl)-2-hydroxy-2-*p*-tolylfuran-3(2H)-ylidene)indolin-2-one (5s): Red solid, 90%. m.p. 194-196□. IR (KBr): 3445, 2025, 1666, 1570, 1543, 1485, 1445, 1407, 1347, 1222, 1183, 1110, 1091, 1055, 1010, 982, 932, 872, 835, 812, 718, 693, 652. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.62 (s, 1H, NH), 9.05 (s, 1H, OH), 7.95 (s, 1H, CH), 7.84 (d, *J* = 7.8Hz, 2H, ArH), 7.65 (d, *J* = 8.4Hz, 2H, ArH), 7.50 (d, *J* = 7.8Hz, 2H, ArH), 7.36 (s, 1H, ArH), 7.20 (d, *J* = 7.8Hz, 1H, ArH), 6.70 (d, *J* = 7.8Hz, 1H, ArH), 2.28 (s, 3H, CH3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.5, 166.6, 157.1, 139.1, 136.3, 134.7, 129.2, 129.0, 128.5, 127.5, 126.5, 125.2, 124.2, 123.8, 123.2, 113.9, 111.6, 110.0, 101.1, 20.7. HRMS (ESI) Calcd. for $C_{25}H_{17}Cl_2NNaO_3$ ($[M+Na]^+$): 472.0478. Found: 472.0485.

3-(5-(4-chlorophenyl)-2-hydroxy-2-p-tolylfuran-3(2H)-ylidene)-5-fluoroindolin-2-one(5t): Red solid, 92%. m.p. 176-178□. IR (KBr): 3615, 3438, 2025, 1655, 1570, 1543, 1481, 1408, 1383, 1351, 1290, 1263, 1243, 1186, 1149, 1112, 1092, 1055, 1011, 992, 938, 866, 834, 810, 698, 654, 625. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.50 (s, 1H, NH), 9.03 (s, 1H, OH), 7.95 (s, 1H,

CH), 7.83 (d, *J* = 7.8Hz, 2H, ArH), 7.55 (d, *J* = 8.4Hz, 2H, ArH), 7.50 (d, *J* = 7.8Hz, 2H, ArH), 7.20 (d, *J* = 7.8Hz, 2H, ArH), 7.09 (d, *J* = 9.6Hz, 1H, ArH), 6.80 (t, *J* = 8.4Hz, 1H, ArH), 6.68(mix, 1H, ArH), 2.28 (s, 3H, CH3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.8, 166.3, 157.2(d, *J* = 119.7Hz), 138.7, 136.8, 136.2, 134.8, 129.2, 129.1, 128.4, 127.5, 125.2, 122.7(d, *J* = 9.0Hz), 114.7(d, *J* = 11.6Hz), 113.3(d, *J* = 24.0Hz), 111.6, 110.9, 109.2(d, *J* = 7.5Hz), 101.1, 20.7. HRMS (ESI) Calcd. for $C_{25}H_{17}ClFNNaO_3$ ($[M+Na]^+$): 456.0773. Found: 456.0775.

Acknowledgment: This work was financially supported by the National Natural Science Foundation of China (Grant No. 21172189) and the Priority Academic Program Development of Jiangsu Higher Education Institutions.

Supporting Information: Crystallographic data **3e** (CCDC 906554), **4a** (CCDC 904920), **5b** (CCDC 904921), and **5l** (CCDC 906832) have been deposited at the Cambridge Crystallographic Database Centre.

References

1. (a) Sundberg, R. J. The Chemistry of Indoles; Academic Press: New York, **1996**; (b) Abdel-Rahman, A. H.; Keshk, E. M.; Hanna, M. A.; El-Bady, Sh. M. *Bioorg. Med. Chem.* **2004**, *12*, 2483-2488; (c) Singh, G. S.; Desta, Z. Y. *Chem. Rev*. **2012**, dx.doi.org/10.1021/cr300135y.

2. (a) Marti, C.; Carreia, E. M. *Eur. J. Org. Chem.* **2003**, 2209; (b) Ashimori, A.; Bachand, B.; Overman, L. E.; Poon, D. J. *J. Am. Chem. Soc.* **199**8, *120*, 6477-6487; (c) Sebahar, P. R.; Williams, R. M. *J. Am. Chem. Soc.* **200**0, *122*, 5666-5667; (d) Tan, B.; Candeias, N. R.; Barbas, C. F. III *Nature. Chem.* **2011**, *3*, 473-474.

3. (a) Zhu, S. -L.; Jia, S. -J.; Zhang, Y. *Tetrahedron* **2007**, *63*, 9365-9372; (b) Kumar, R. S.; Perumal, S. *Tetrahedron Lett.* **2007**, *48*, 7164-7168; (c) Redkin, R. Gr.; Shemchuk, L. A.; Chernykh, V. P.; Shishkin, O. V.; Shishkina, S. V. *Tetrahedron* **2007**, *63*, 11444-11450.

4. (a) Lo, M. M. C.; Neumann, C. S.; Nagayama, S.; Perlstein, E. O.; Schreiber, S. L. *J. Am. Chem. Soc.* **2004**, *12*6, 16077–16086; (b) Galliford, C. V.; Scheidt, K. A. *Angew. Chem. Int. Ed.* 2007, *46*, 8748–8758; (c) Chen, X.; Wei, Q.; Luo, S.; Xiao, H.; Gong, L. *J. Am. Chem. Soc.* **2009**, *131*, 13819–13825.

5. (a) Robertson, D. W.; Krushinski, J. H.; Pollock, G. D.; Wilson, H.; Kauffman, R. F.; Hayes, J. S. *J. Med. Chem.* **1987**, *30*, 824-829; (b) Jiang, T.; Kuhen, K. L.; Wolff, K.; Yin, H.; Bieza, K.; Caldwell, J.; Bursulaya, B.; Wu, T.

H.; He, Y. *Bioorg. Med. Chem. Lett.* **2006**, *16*, 2105-2108.

6. (a) Alper, P. B.; Meyers, C.; Lerchner, A.; Siegel, D. R.; Carreira, E. M. *Angew.Chem., Int. Ed.* **1999**, *38*, 3186-3189; (b) Wood, J. L.; Holubec, A. A.; Stoltz, B. M.; Weiss, M. M.; Dixon, J. A.; Doan, B. D.; Shamji, M. F.; Chen, J. M.; Heffron, T. P. J. *Am. Chem. Soc.* **1999**, *121*, 6326- ; (c) Lerchner, A.; Carreira, E. M. *J. Am. Chem. Soc*. **2002**, *124*, 14826-14827; (d) Marti, C.; Carreira, E. M. *J. Am. Chem. Soc*. **2005**, *127*, 11505-11515.
7. (a) Franke, A. *Justus Liebigs Ann. Chem.* **1978**, *71*, 7; (b) Bennett, *G.;* Mason, R. B.; Shapiro, M. J. *J. Org. Chem.* **1978**, *43*, 4383-4385; (c) Joshi, K. C.; Jain, R.; Amra, S. *Heterocycles*, **1987**, *26*, 1835-1839; d) Muthusamy, S.; Gunanathan, C. *Synlett* **2003**, 1599-1602; (d) Reddy, B. V. S.; Rajasekaran, T.; Karthik, G.; Rao, T. P. *Tetrahedron Lett.* **2012**, *53*, 3416-3420.
8. (a) Chiericato, M.; Dalla Croce, P.; Licandro, E. *J. Chem. Soc. Perkin Trans. I.* **1979**, 211-213; (b) S. R. Yong, A. T. Ung, S. G. Pyne, B. W. Skelton, A. H. White, *Tetrahedron* **2007**, *63*, 1191-1199;
9. (a) Eberle, M. K.; Kahle, G. G.; Shapiro, M. J. *J. Org. Chem.* **1982,** *47*, 2210-2212; (b) H. Yu, Y. Liu, H. Zhang, J. Chen, H. Deng, M. Shao, Z. Ren, W. Cao, *Tetrahedron* **2010**, *66*, 2598-2601.
10. (a) Morales-Ríos, M. S.; González-Juárez, D. E.; Rivera-Becerril, E.; Suárez-Castillo, O. R.; Joseph-Nathana, P. *Tetrahedron* **2007**, *63*, 7702–7707; (b) Gonzáalez-Juáarez, D. E.; García-Váazquez, J. B.; Zúniga-García, V.; Trujillo-Serrato, J. J.; Suárez-Castillo, O. R.; Joseph-Nathan, P.; Morales-Ríos, M. S. *Tetrahedron* **2012**, *68*, 7187-7195.
11. (a) Shanmugam, P.; Vaithiyanathan, V.; Viswambharan, B. *Tetrahedron* **2006**, *62*, 4342- ; (b) Jaegli, S.; Vors, J. P.; Neuville, L.; Zhu, J. P. *Tetrahedron*, **2010,** *66*, 8911-8921; (c) Lingam, K. A. P.; Mandal, A. B.; Shanmugam, P. *Tetrahedron Lett.* **2011**, *52*, 3610–3613; (d) Ivashkin, P.; Couve-Bonnaire, S.; Jubault, P.; Pannecoucke, X.; *Org. Lett.*, **2012**, *14* (9), 2270–2273.
12. Pesciaioli, F.; Righi, P.; Mazzanti, A.; Bartoli, G.; Bencivenni, G. *Chem. Eur. J*. **2011**, *17*, 2842–2845.
13. Wang, Q. F.; Song, X. K.; Chen, J.; Yan, C. G. *J. Comb. Chem.* **2009,** *11*, 1007–1010.
14. (a) Kröhnke, F.; Zecher, W. *Angew. Chem. Int. Ed.* **1962**, *1*, 626; (b) Kröhnke, F. *Angew. Chem. Int. Ed*. **1963**, *2*, 225.
15. (a) Yan, C. G.; Cai, X. M.; Wang, Q. F.; Wang, T. Y.; Zheng, M. *Org. Biomol. Chem.* **2007**, *5*, 945-951; (b) Yan, C. G.; Song, X. K.; Wang, Q. F.; Sun, J.; Siemeling, U.; Bruhn, C. *Chem. Commun.* **2008**, 1440-1442; (c) Yan, C. G.; Wang, Q. F.; Song, X. K.; Sun, J. *J. Org. Chem.* **2009**, *74*, 710-718; (d) Wang, Q. F.; Hou, H.; Hui, L.; Yan, C. G. *J. Org. Chem.* **2009**, ***74***, 7403-7408.

16. (a) Papageorgiou, C. D.; Ley, S. V.; Gaunt, M. J. *Angew. Chem., Int. Ed.* **2003**, *42*, 828–831; (b) Papageorgiou, C. D.; Cubillo de Dios, M. A.; Ley, S. V.; Gaunt, M. J. *Angew. Chem., Int. Ed.* **2004**, *43*, 4641–4644; (c) Vanecko, J. A.; Wan, H.; West, F. G. *Tetrahedron*, **2006**, *62*, 1043–1062; (d) Bhattacharjee, S. S.; Ila, H.; Junjappa, H. *Synthesis* **1982**, 301–303; (e) Jończyk, A.; Konarska, A. *Synlett* **1999**, 1085–1087; (f) Joæczyk, A.; Sucholbiak, D.; Jończyk, A. *Eur. J. Org. Chem.* **2005**, 925–933.

17. (a) Nyerges, M.; Gajdics, L.; Szöllösy, A.; Töke, L.; *Synlett* **1999**, 111-113; (b) I. Fejes, L. Toke, M. Nyerges, and C. S. Pak, *Tetrahedron*, **2000**, *56*, 639-644; (c) I. Fejes, M. Nyerges, A. Szollosy, G. Blasko, and L. Toke, *Tetrahedron*, **2001**, ***57***, 1129-1137.

18. Serov, A. B.; kartsev, V. G.; Aleksandrov, Yu, A.; Dolgushin, F. M. *Russ. Chem. Bull.* **2005**, *54*, 2432-2436.

19. El-Ahl, A. A. S.; *Pol. J. Chem.* **1997**, *71*, 27-31.

20. (a) Sun, Y.; Sun, J.; Yan, C. G. *Tetrahedron Lett.* **2012**, 53, 3647; (b) Han, Y.; Wu, Q.; Sun, J.; Yan, C. G. *Tetrahedron*, **2012**, *68*, 8539; (c) Hui, L.; Li, H. Y.; Yan, C. G. *Eur. J. Org. Chem.* **2012**, 315; (d) Sun, J.; Sun, Y.; Gao, H.; Yan, C. G *Eur. J. Org. Chem.* **2012**, 1976.

21. (a) Aggarwal, V. K.; Alsono, E.; Fang, G.; Ferrara, M.; Hynd, G.; Porcelloni, M. *Angew. Chem., Int. Ed.* **2001**, *40*, 1433–1435; (b) Wang, Q. G.; Deng, X. M.; Zhu, B. H.; Ye, L. W.; Sun, X. L.; Li, C. Y.; Zhu, C. Y.; Shen, Q.; Tang, T. *J. Am. Chem. Soc.* **2008**, *130*, 5408–5409; (c) Cao, W. G.; Zhang, H.; Chen, J.; Deng, H. M.; Shao, M.; Lei, L. Qian, J. X.; Zhu, Y. *Tetrahedron* **2008**, *64*, 6670–6674.

22. (a) Autrey, R. L.; Tahk, F. C. *Tetrahedron*, **1967**, *23*, 901-917; (b) Kloek, C.; Jin, X.; Choi, K.; Khosla, C.; Madrid, P. B.; Spencer, A.; Raimundo, B. C.; Boardman, P.; Lanza, G.; Griffin, J. H.; *Bioorg. Med. Chem. Lett.* **2011**, *21*, 2692–2696; (c) Tan, B.; Candeias, N. R.; Barbas, C. F. III *Nature. Chem.* **2011**, *3*, 473-474.

23. (a) Wang, L.; Zhang, Y.; Hu, H. Y.; Fun, H. K.; Xu, J. H. *J. Org. Chem.* **2005**, *70*, 3850-3858; (b) Shanmugam, P.; Viswambharan, B. Madhavan, S. *Org. Lett.* **2007**, *9*, 4095-4098; (c) Han, Y.; Sun, Y.; Sun, J.; Yan, C. G. *Tetrahedron*, **2012**, *68*, 8256;

24. (a) Rehn, S.; Bergman, J. *Tetrahedron* **2005**, *61*, 3115-3123; (b) Muthusamy, S.; Gunanathan, C. *Synlett* **2002**, 1783-1787; (c) Shanthi, G.; Perumal, G. T. *Tetrahedron Lett.* **2009**, *50*, 3959–3962.

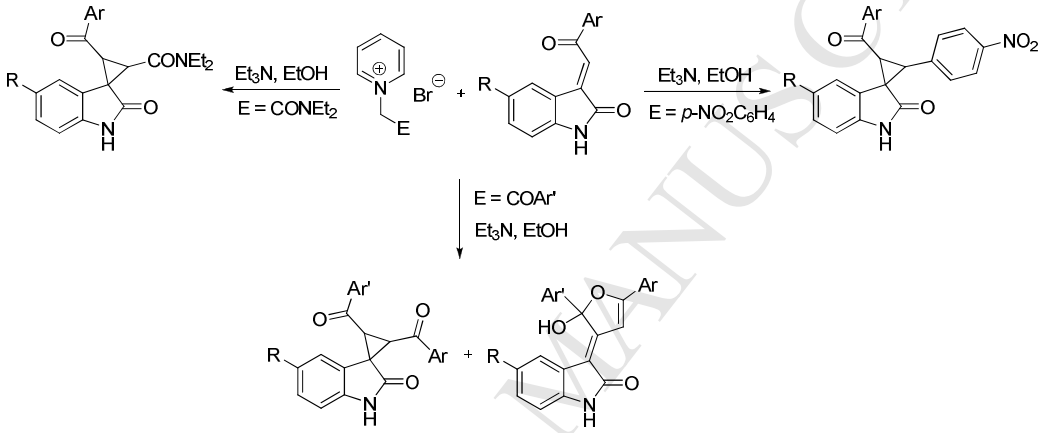
25. Higashiyama, K.; Nagase, H.; Yamaguchi, R.; Kawai, K.; Otomasu, H. *Chem. Pharm. Bull.* **1985**, *33*, 544-550.

26. (a) Sarkunam, K.; Nallu, M. *J. Heterocyclic Chem*. **2005**, *42*, 5-11; (b) Murphy, J. A.; Garnier, J.; Park, S. R.; Schoenebeck, F.; Zhou, S. Z.; Turner, A. T. *Org. Lett.*, **2008**, *10*, 1227–1230; (c) Belei, D.; Abuhaie, C.; Bicu, E.; Jones, P. G.; Hopf, H.; Birsa, L. M. *Synlett*, **2012**, 545-548.x.x.

Graphic abstract:

Molecular diversity of cycloaddition reactions of functionalized pyridinium salts with 3-phenacylideneoxindoles

Qin Fu, Chao-Guo Yan



Molecular diversity of cycloaddition reactions of the functionalized pyridinium salts with 3-phenacylideneoxindoles

Qin Fu, Chao-Guo Yan*

Supporting Information

General Experimental Methods and Characterization of compounds **2-15**

1H NMR and 13C NMR of the preparaed compounds **16-53**

X-Ray Crystallographic Data: CIF in separate file.

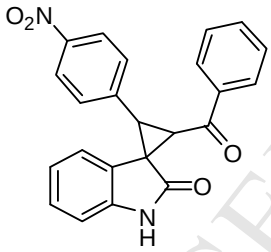
Crystallographic data **3e** (CCDC 906554), **4a** (CCDC 904920), **5b** (CCDC 904921), and **5l** (CCDC 906832) have been deposited vat the Cambridge Crystallographic Database Centre and is available on request from the Director, CCDC, 12 Union Road, Cambridge, CB2 1EZ, UK (http//www.ccdc.cam.ac.uk).

Experimental Section

All reagents and solvents were commercial available with analytical grade and used as received. All evaporations of organic solvents were carried out with a rotary evaporator in conjunction with a water aspirator. Melting points were taken on a hot-plate microscope apparatus and were uncorrected. ^{1}H and ^{13}C NMR spectra were recorded with a Bruker AV-600 instrument. IR spectra were obtained on a Bruker Tensor 27 spectrometer (KBr disc). HRMS were measured at AB 5800 MALDI-TOF/TOF instrument. X-ray data were collected on a Bruker Smart APEX-2 diffractometer. Pyridinium salts were prepared by heating pyridine with *p*-nitrobenzyl bromide, *α*-phenacyl bromide, and *N*,*N*-diethyl chloroacetamide in acetonitrile according to the published procedure.

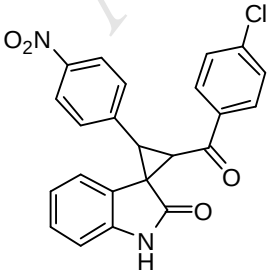
1. General procedure for the preparation of spiro[cyclopropane-1,3'-indolines] 3a-3j from the reactions of pyrididinium salts with 3-phenacylideneoxindoles: A mixture of *N*-*p*-nitrobenzylpyridinium bromide or *N*-diethylcarbamoylmethylpyridinium chloride (1.1 mmol) 3-phenacylideneoxindoles (1.0 mmol) and triethylamine (0.2 mmol) in ethanol (15mL) was stirred at about 50°C for six hours. The resulting precipitates were collected by filtration, which were recrystallized in a mixture of chloroform and ethanol to give pure product for analysis.

2-benzoyl-3-(4-nitrophenyl)spiro[cyclopropane-1,3'-indolin]-2'-one (3a):



White solid, 78%. m.p. 216-218°C. IR (KBr): 3422, 3081, 1700, 1621, 1600, 1515, 1469, 1345, 1281, 1108, 1017, 975, 847. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.25 (s, 1H, NH), 8.18 (d, *J* = 8.4Hz, 2H, ArH) , 7.96 (d, *J* = 7.8Hz, 2H, ArH), 7.58 (t, *J* = 7.2Hz, 1H, ArH), 7.53 (d, *J* = 8.4Hz, 2H, ArH), 7.44 (t, *J* = 7.2Hz, 2H, ArH), 7.25 (d, *J* = 7.8Hz, 1H, ArH), 7.19 (t, *J* = 7.8Hz, 1H, ArH), 6.99 (t, *J* = 7.2Hz, 1H, ArH), 6.84 (d, *J* = 7.2Hz, 1H, ArH), 4.30 (d, *J* = 7.8Hz, 1H, CH), 4.17 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 192.3, 173.9, 147.2, 141.3, 141.2, 136.7, 134.0, 130.3, 128.9, 128.5, 128.1, 125.6, 123.3, 122.6, 122.3, 110.2, 42.0, 41.0, 38.3. HRMS (ESI) Calcd. for $C_{23}H_{16}N_2NaO_4$ ($[M+Na]^+$): 407.1002. Found: 407.1003.

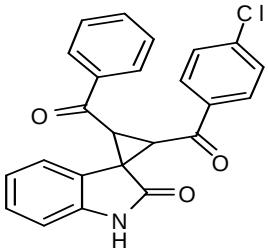
2-(4-chlorobenzoyl)-3-(4-nitrophenyl)spiro[cyclopropane-1,3'-indolin]-2'-one (3b):



White solid, 76%. m.p. 222-224°C. IR (KBr): 3444, 1721, 1671, 1629, 1525, 1469, 1047, 1349, 1191, 1089, 1012, 842. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.19 (d, *J* = 9.0Hz, 2H, ArH), 7.90 (d, *J* = 9.0Hz, 2H, ArH), 7.81 (s, 1H, NH), 7.52 (d, *J* = 8.4Hz, 2H, ArH), 7.41 (d, *J* = 8.4Hz, 2H, ArH), 7.12 (t, *J* = 7.2Hz, 2H, ArH), 7.00 (t, *J* = 7.8Hz, 1H, ArH), 6.88 (d, *J* = 7.8Hz, 1H, ArH), 4.23 (d, *J* = 7.8Hz, 1H, CH), 4.15 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.0,

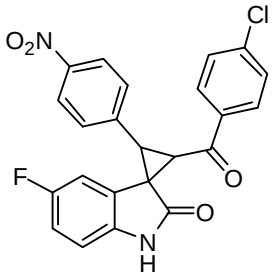
172.9, 147.3, 140.7, 134.8, 130.2, 129.8, 129.3, 128.3, 125.3, 123.4, 122.8, 122.3, 110.0, 41.7, 40.9, 38.4. HRMS (ESI) Calcd. for $C_{23}H_{15}ClN_2NaO_4$ ([M+Na]$^+$): 441.0613. Found: 441.0612.

2-benzoyl-5'-fluoro-3-(4-nitrophenyl)spiro[cyclopropane-1,3'-indolin]-2'-one (3c):



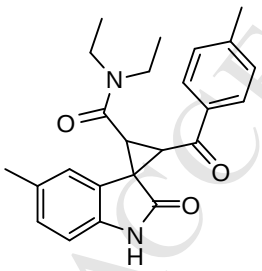
White solid, 65%. m.p. 204-206°C. IR (KBr): 3438, 3383, 1727, 1667, 1637, 1636, 1601, 1522, 1481, 1351, 1325, 1221, 1178, 1147, 1110. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.19 (d, *J* = 7.8Hz, 2H, ArH), 7.99 (d, *J* = 7.8Hz, 3H, ArH, NH), 7.61 (t, *J* = 7.2Hz, 1H, ArH), 7.53 (d, *J* = 8.4Hz, 2H, ArH), 7.48 (t, *J* = 7.2Hz, 2H, ArH), 7.08 (d, *J* = 6.6Hz, 1H, ArH), 6.91 (t, *J* = 8.4Hz, 1H, ArH), 6.79-6.77 (m, 1H, ArH), 4.33 (d, *J* = 7.8Hz, 1H, CH), 4.15 (d, *J* = 7.8Hz, 1H, CH) . ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.8, 173.0, 158.5 (d, *J* = 239.0Hz) , 147.4, 140.5, 136.7, 136.5, 134.2, 130.3, 129.0, 128.5, 127.1 (d, *J* = 9.0Hz), 122.4, 114.6 (d, *J* = 25.1Hz), 110.8 (d, *J* = 26.0Hz), 110.4 (d, *J* = 8.0Hz), 41.9, 40.9, 39.0. HRMS (ESI) Calcd. for $C_{23}H_{15}FN_2NaO_4$ ([M+Na]$^+$): 425.0908. Found: 425.0906.

2-(4-chlorobenzoyl)-5'-fluoro-3-(4-nitrophenyl)spiro[cyclopropane-1,3'-indolin]-2'-one (3d):



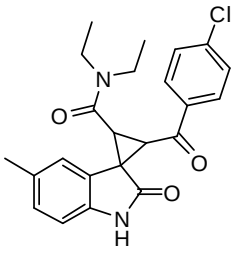
White solid, 70%. m.p. 236-238°C. IR (KBr): 3448, 1726, 1675, 1637, 1625, 1601, 1520, 1471, 1340, 1229, 1178, 1080, 1110, 857. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.27 (s, 1H, NH), 8.13 (d, *J* = 8.4Hz, 2H, ArH), 7.94 (d, *J* = 8.4Hz, 2H, ArH), 7.63 (d, *J* = 8.4Hz, 2H, ArH), 7.56 (d, *J* = 8.4Hz, 2H, ArH), 7.31 (dd, J_1= 2.4Hz, J_2 = 8.4Hz, 1H, ArH), 7.18 (dt, J_1= 1.8Hz, J_2 = 8.4Hz, 1H, ArH), 7.05 (dd, J_1= 3.6Hz, J_2 = 7.8Hz, 1H, ArH), 4.54 (d, *J* = 7.8Hz, 1H, ArH), 4.28 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.0, 173.5, 159.6 (d, *J* = 239.6Hz), 141.4, 141.0, 137.8, 135.2, 134.7, 130.5, 130.2, 129.8, 125.9 (d, *J* = 9.0Hz), 116.0 (d, *J* = 23.7Hz), 111.7 (d, *J* = 8.1Hz), 111.4 (d, *J* = 26.3Hz), 41.7, 40.3, 39.0. HRMS (ESI) Calcd. for $C_{23}H_{14}FClN_2NaO_4$ ([M+Na]$^+$): 483.1470. Found: 483.1474.

***N*,*N*-diethyl-5'-methyl-3-(4-methylbenzoyl)-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carbox amide (3e):**



White solid, 72%. m.p. 174-176°C. IR (KBr): 3463, 2975, 1723, 1693, 1635, 1488, 1457, 1422, 1321, 1208, 1180, 1144, 1079, 1029, 985, 822. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.37 (s, 1H, NH), 7.72 (d, *J* = 7.8Hz, 2H, ArH), 7.14 (s, 2H, ArH), 7.13 (s, 1H, ArH), 7.05 (d, *J* = 7.8Hz, 1H, ArH), 6.78 (d, *J* = 7.8Hz, 1H, ArH), 3.97 (d, *J* = 7.8Hz, 1H, CH), 3.56-3.50 (m, 2H, 2CH), 3.24-3.15 (m, 3H, CH, CH_2), 2.33 (s, 3H, CH_3), 2.32 (s, 3H, CH_3), 1.03 (t, *J* = 7.2Hz, 3H, CH_3), 0.95 (t, *J* = 7.2Hz, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.6, 173.8, 164.5, 144.3, 139.0, 133.9, 132.0, 129.4, 128.7, 128.5, 125.0, 122.6, 110.0, 42.2, 40.8, 39.7, 38.7, 35.0, 21.6, 21.2, 14.0, 13.1. HRMS (ESI) Calcd. for $C_{24}H_{26}N_2NaO_3$ ([M+Na]$^+$): 413.2662. Found: 413.2658.

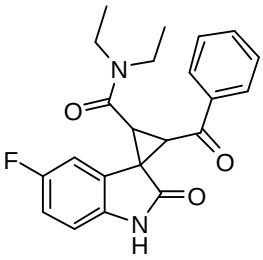
3-(4-chlorobenzoyl)-*N*,*N*-diethyl-5'-methyl-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carbox amide (3f):



White solid, 88%. m.p. 160-162°C. IR (KBr):3443, 3233, 2972, 1720,

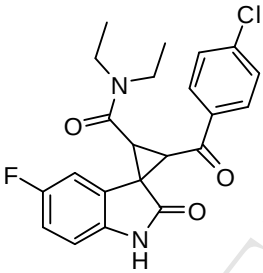
1628, 1591, 1488, 1424,1318, 1201, 1145, 1086, 1034, 911, 858. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.09 (s, 1H, NH), 7.76 (d, *J* = 7.8Hz, 2H, ArH), 7.33 (d, *J* = 7.8Hz, 2H, ArH), 7.15 (s, 1H, ArH), 7.07 (d, *J* = 7.8Hz, 1H, ArH), 6.80 (d, *J* = 7.8Hz, 1H, ArH), 3.95 (d, *J* = 7.8Hz, 1H, CH), 3.58-3.52 (m, 2H, 2CH), 3.24-3.16 (m, 3H, CH, CH_2), 2.32 (s, 3H, CH_3), 1.04 (t, *J* = 7.2Hz, 3H, CH_3), 0.96 (t, *J* = 7.2Hz, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 189.9, 173.6, 164.2, 139.9, 139.0, 134.6, 132.3, 129.8, 129.0, 128.9, 124.7, 122.7, 110.1, 42.2, 40.8, 39.5, 38.6, 35.0, 21.2, 14.0, 13.2. HRMS (ESI) Calcd. for $C_{23}H_{23}ClN_2NaO_3$ ($[M+Na]^+$): 433.1289. Found: 433.1290.

2-benzoyl-*N*,*N*-diethyl-5'-fluoro-2'-oxospiro[cyclopropane-1,3'-indoline]-3-carboxamide (3g):



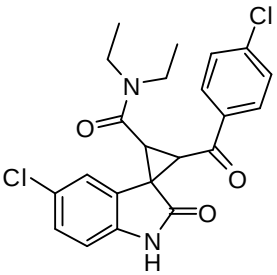
White solid, 69%. m.p. 200-202°C. IR (KBr): 3366, 3276, 3068, 2972, 2934, 1708, 1637, 1484, 1423, 1322, 1223, 1156, 1089, 1026, 959, 832. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.85 (s, 1H, NH), 7.80 (d, *J* = 7.8Hz, 2H, ArH), 7.49 (t, *J* = 7.8Hz, 1H, ArH), 7.35 (t, *J* = 7.8Hz, 1H, ArH), 7.19 (dd, J_1 = 8.4Hz, J_2= 2.4Hz, 1H, ArH), 6.98 (td, J_1 = 8.4Hz, J_2= 2.4Hz, 1H, ArH) , 6.88 (dd, J_1 = 7.8Hz, J_2 = 1.8Hz, 1H, ArH), 4.03 (d, *J* = 7.8 Hz, 1H, CH), 3.58 (d, *J* = 7.8Hz, 1H, CH), 3.52-3.46 (m, 1H, CH), 3.32-3.27 (m, 1H, CH), 3.26-3.22 (m, 2H, CH_2) , 1.05 (t, *J* = 7.2Hz, 3H, CH_3), 0.97 (t, *J* = 7.2Hz, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.4, 173.8, 164.0, 159.8, 158.2, 137.5, 136.1, 133.6, 128.8, 128.4, 126.6, 126.5, 115.0, 114.8, 111.1, 111.0, 110.5, 110.3, 42.3, 40.9, 40.1, 38.9, 35.1, 14.2, 13.2. HRMS (ESI) Calcd. for $C_{22}H_{21}FN_2NaO_3$ ($[M+Na]^+$): 403.1428. Found: 403.1432.

3-(4-chlorobenzoyl)-*N*,*N*-diethyl-5'-fluoro-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carboxa mide (3h):



White solid, 54%. m.p. 230-232°C. IR (KBr): 3451, 2970, 1731, 1634, 1592, 1485, 1419, 1223, 1183, 1148, 1091, 1016, 957, 822. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.34 (s, 1H, NH), 7.75 (d, *J* = 7.8Hz, 2H, ArH), 7.35 (d, *J* = 7.8Hz, 2H, ArH), 7.20 (d, *J* = 8.4Hz, 1H, ArH), 7.00 (t, *J* = 8.4Hz, 1H, ArH), 6.87 (dd, J_1 = 7.8Hz, J_2 = 3.6Hz, 1H, ArH), 3.99 (d, *J* = 7.8Hz, 1H, CH), 3.57 (d, *J* = 7.8Hz, 1H, CH), 3.52-3.47 (m, 1H, CH), 3.33-3.29 (m, 1H, CH), 3.27-3.24 (m, 2H, CH_2), 1.07 (t, *J* = 7.2Hz, 3H, CH_3), 1.00 (t, *J* = 7.2Hz, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 189.4, 173.5, 163.9, 159.1 (d, *J* = 239.6 Hz) , 140.1, 137.3, 134.4, 129.7, 129.1, 126.3 (d, *J* = 9.6Hz), 115.0 (d, *J* = 23.9Hz), 110.9 (d, *J* = 7.8Hz), 110.5 (d, *J* = 26.3Hz) , 40.3, 41.0, 39.9, 38.8, 35.1, 14.2, 13.2. HRMS (ESI) Calcd. for $C_{22}H_{20}ClFN_2NaO_3$ ($[M+Na]^+$): 437.1039. Found: 437.1041.

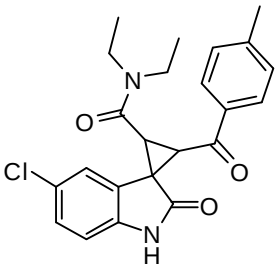
5'-chloro-3-(4-chlorobenzoyl)-*N*,*N*-diethyl-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carboxa mide (3i):



White solid, 82%. m.p. 242-244°C. IR (KBr): 3420, 2982, 1703, 1631, 1591, 1480, 1420, 1322, 1270, 1208, 1141, 1092, 825. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.58 (s, 1H, NH), 7.74 (d, *J* = 8.4Hz, 2H, ArH), 7.39 (s, 1H, ArH), 7.35 (d, *J* = 8.4Hz, 2H, ArH), 7.26 (d, *J* = 8.4Hz, 1H, ArH), 6.88 (d, *J* = 8.4Hz, 1H, ArH), 4.00 (d, *J* = 7.8Hz, 1H, CH), 3.57-3.52 (m, 2H, 2CH), 3.28-3.19 (m, 3H, CH,

CH_2), 1.06(t, *J* = 7.2Hz, 3H, CH_3), 0.99 (t, *J* = 7.2Hz, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 189.3, 173.3, 163.8, 130.2,139.9, 134.4,129.8, 129.2, 128.6, 128.3, 126.4, 122.7, 111.3, 42.3, 41.1, 39.8, 38.4, 35.1, 14.2, 13.2. HRMS (ESI) Calcd. for $C_{22}H_{20}Cl_2N_2NaO_3$ ([M+Na]$^+$): 453.0743. Found: 453.0745.

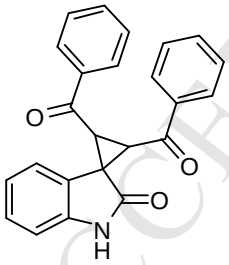
5'-chloro-*N*,*N*-diethyl-3-(4-methylbenzoyl)-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carbox amide (3j):



White solid, 80%. m.p. 166-168°C. IR (KBr): 3460, 2981, 2026, 1703, 1633, 1420, 1322, 1271, 1208, 1141, 1085, 823. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.44 (s, 1H, NH), 7.69 (brs, 2H, ArH), 7.40 (s, 1H, ArH), 7.22 (brs, 1H, ArH), 7.14 (brs, 2H, ArH), 6.83 (brs, 1H, ArH), 4.02 (d, *J* = 6.6Hz, 1H, CH), 3.58 (d, *J* = 6.6Hz, 1H, CH), 3.53 (brs, 1H, CH), 3.22 (brs, 3H, CH, CH_2), 2.33 (s, 3H, CH_3), 1.05 (brs, 3H, CH_3), 0.96 (brs, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.1, 173.5, 163.9, 144.7, 139.8, 133.6, 129.5, 128.5, 128.4, 128.1, 126.7, 122.6, 111.2, 42.3, 41.0, 40.1, 38.5, 35.2, 21.7, 13.1, 13.2. HRMS (ESI) Calcd. for $C_{23}H_{23}ClN_2NaO_3$ ([M+Na]$^+$): 433.1289. Found: 433.1292.

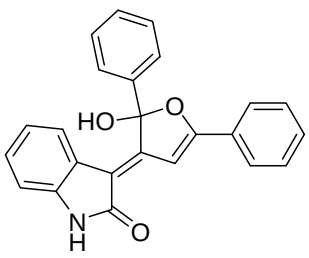
2. General procedure for the preparation of spiro[cyclopropane-1,3'-indolines] 4a-4q and 3-furan-3(2H)-ylidene)indolin-2-ones 5a-5q from the reactions of pyrididinium salts with 3-phenacylideneoxindoles: A mixture of *N*-phenacylpyridinium bromide or *N*-*p*-chlorophenacylpyridinium chloride (1.1 mmol) 3-phenacylideneoxindoles (1.0 mmol) and triethylamine (0.2 mmol) in ethanol (15mL) was stirred at 50°C for six hours. The resulting precipitates were collected by filtration, which was bjected to thin-layer chromatography with a mixture of ethyl acetate and light petroleum (V/V = 2:1) to give the pure product for analysis.

(2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis(phenylmethanone) (4a):



White solid, 54%. m.p. 176-178°C. IR (KBr): 3256, 1719, 1673, 1620, 1595, 1465, 1400, 1308, 1219, 1085, 1020, 936, 801. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.53 (s, 1H, NH), 7.96 (d, *J* = 7.8Hz, 2H, ArH), 7.82 (d, *J* = 7.8Hz, 2H, ArH), 7.53 (t, *J* = 7.2Hz, 1H, ArH), 7.48 (t, *J* = 7.2Hz, 1H, ArH), 7.40 (t, *J* = 7.8Hz, 2H, ArH), 7.35-7.32 (m, 3H, ArH), 7.24 (t, *J* = 7.2Hz, 1H, ArH), 7.03 (t, *J* = 7.2Hz, 1H, ArH), 6.93 (d, *J* = 7.8Hz, 1H, ArH), 4.41 (d, *J* = 7.8Hz, 1H, CH), 4.17 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 192.1, 190.5, 173.6, 141.7, 136.7, 136.1, 133.8, 133.6, 128.8, 128.7, 128.6, 128.4, 124.3, 122.7, 122.5, 110.7, 41.3, 39.6, 38.7. HRMS (ESI) Calcd. for $C_{24}H_{17}NaNO_3$ ([M+Na]$^+$): 390.1101. Found: 390.1103.

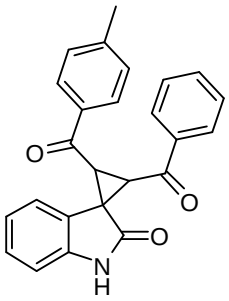
3-(2-hydroxy-2,5-diphenylfuran-3(2H)-ylidene)indolin-2-one (5a):



Yellow solid, 28%. m.p. 134-136°C. IR (KBr): 3445, 3252, 1633, 1616, 1545, 1483, 1389, 1348, 1265, 1211, 1180, 1118, 969, 829. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.46 (s, 1H, NH), 8.91 (s, 1H, OH), 7.97 (s, 1H, ArH), 7.79 (d, *J* = 7.2Hz, 2H, ArH), 7.63 (d, *J* = 6.0Hz, 2H, ArH), 7.52-7.47 (m, 3H, ArH), 7.39-7.34 (m, 3H,

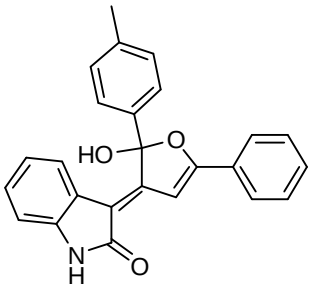
ArH), 7.29 (s, 1H, ArH), 6.93 (t, *J* = 6.6Hz, 1H, ArH), 6.69 (d, *J* = 7.8Hz, 1H, ArH), 6.55 (t, *J* = 7.8Hz, 1H, ArH). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.8, 166.7, 155.3, 140.5, 138.1, 136.1, 133.3, 131.4, 129.0, 128.4, 126.5, 125.3, 122.8, 122.7, 120.0, 117.8, 111.9, 108.7, 100.6. HRMS (ESI) Calcd. for $C_{24}H_{17}NNaO_3$ ([M+Na]$^+$): 390.1101. Found: 390.1101.

2-benzoyl-3-(4-methylbenzoyl)spiro[cyclopropane-1,3'-indolin]-2'-one (4b):



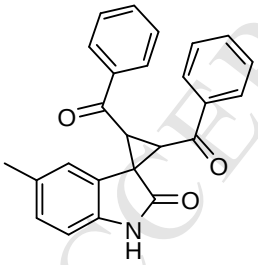
White solid, 48%. m.p. 192-194°C. IR (KBr): 3281, 3021, 1726, 1670, 1601, 1470, 1406, 1306, 1218, 1094, 1019, 921, 832. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.52 (s, 1H, NH), 7.96 (d, *J* = 7.2Hz, 2H, ArH), 7.71 (d, *J* = 7.2Hz, 2H, ArH), 7.53 (t, *J* = 6.6Hz, 1H, ArH), 7.40 (t, *J* = 7.2Hz, 2H, ArH), 7.32 (d, *J* = 7.2Hz, 1H, ArH), 7.23 (t, *J* = 7.8Hz, 1H, ArH), 7.13 (t, *J* = 7.8Hz, 2H, ArH), 7.02 (t, *J* = 7.2Hz, 1H, ArH), 6.91 (d, *J* = 7.2Hz, 1H, ArH), 4.40 (d, *J* = 7.8Hz, 1H, CH), 4.15 (d, *J* = 7.8Hz, 1H, CH), 2.31 (s, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 192.2, 190.1, 173.6, 144.6, 141.6, 136.8, 133.8, 129.5, 128.8, 128.7, 128.6, 128.5, 128.4, 124.4, 122.7, 122.5, 110.6, 41.3, 39.7, 38.8, 21.6. HRMS (ESI) Calcd. for $C_{25}H_{19}NNaO_3$ ([M+Na]$^+$): 404.1257. Found: 404.1260.

3-(2-hydroxy-5-phenyl-2-p-tolylfuran-3(2H)-ylidene)indolin-2-one (5b):



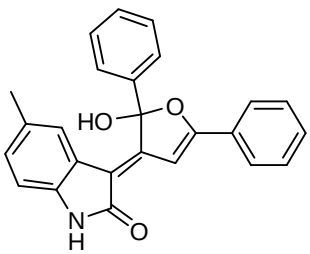
Yellow solid, 31%. m.p. 172-174°C. IR (KBr): 3359, 1670, 1613, 1542, 1489, 1464, 1351, 1255, 1205, 1176, 1149, 1099, 1061, 957, 830. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.44 (s, 1H, NH), 8.83 (s, 1H, OH), 7.94 (s, 1H, ArH), 7.78 (d, *J* = 7.2Hz, 2H, ArH), 7.50-7.47 (m, 5H, ArH), 7.33 (d, *J* = 7.8Hz, 1H, ArH), 7.17 (d, *J* = 7.2Hz, 2H, ArH), 6.94 (t, *J* = 6.6Hz, 1H, ArH), 6.69 (d, *J* = 7.2Hz, 1H, ArH), 6.57 (t, *J* = 7.2Hz, 1H, ArH), 2.26 (s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.9, 166.6, 155.6, 140.5, 138.3, 135.3, 131.4, 129.0, 128.9, 128.7, 127.1, 127.0, 126.4, 125.3, 124.3, 121.8, 120.1, 114.8, 108.8, 100.6, 20.7. HRMS (ESI) Calcd. for $C_{25}H_{19}NNaO_3$ ([M+Na]$^+$): 404.1257. Found: 404.1258.

(5'-methyl-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis(phenylmethanone) (4c):



White solid, 56%. m.p. 194-196°C. IR (KBr): 3202, 1712, 1674, 1625, 1593, 1486, 1448, 1331, 1218, 1082, 1024, 941, 817. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.71 (s, 1H, NH), 7.95 (d, *J* = 7.2Hz, 2H, ArH), 7.80 (d, *J* = 7.2Hz, 2H, ArH), 7.52 (t, *J* = 7.2Hz, 1H, ArH), 7.46 (t, *J* = 7.8Hz, 1H, ArH), 7.39 (t, *J* = 7.8Hz, 2H, ArH), 7.32 (t, *J* = 7.8Hz, 2H, ArH), 7.11 (s, 1H, ArH), 7.03 (d, *J* = 7.8Hz, 1H, ArH), 6.81 (d, *J* = 7.8Hz, 1H, ArH), 4.38 (d, *J* = 7.8Hz, 1H, CH), 4.12 (d, *J* = 7.8Hz, 1H, CH), 2.31 (s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 191.9, 191.0, 172.2, 140.2, 136.3, 135.9, 134.0, 133.6, 130.4, 129.1, 128.9, 128.8, 128.0, 127.9, 124.0, 122.6, 109.8, 40.4, 38.5, 38.4, 20.8. HRMS (ESI) Calcd. for $C_{25}H_{19}NNaO_3$ ([M+Na]$^+$): 404.1257. Found: 404.1260.

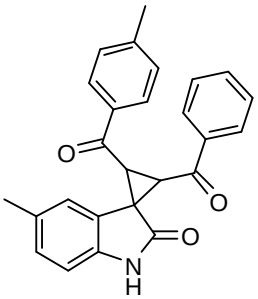
3-(2-hydroxy-2,5-diphenylfuran-3(2H)-ylidene)-5-methylindolin-2-one (5c):



Yellow solid, 30%. m.p. 178-180°C. IR (KBr): 3404, 3196, 1658, 1614, 1580, 1484, 1449, 1386, 1324, 1255, 1203, 1147, 1002, 935, 816. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.35 (s, 1H, NH), 8.88 (s, 1H, OH), 7.97 (s, 1H, ArH), 7.79 (s, 2H, ArH), 7.64 (s, 2H,

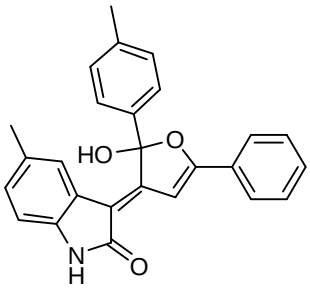
ArH), 7.50 (s, 3H, ArH), 7.37 (s, 3H, ArH), 7.16 (s, 1H, ArH), 6.75 (s, 1H, ArH), 6.59 (s, 1H, ArH), 1.97 (s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 169.0, 166.4, 155.3, 138.3, 138.2, 131.3, 129.0, 128.9, 128.3, 128.2, 127.5, 126.4, 125.4, 125.1, 121.7, 115.1, 111.1, 108.4, 100.6, 20.8. HRMS (ESI) Calcd. for $C_{25}H_{19}NNaO_3$ ([M+Na]$^+$): 404.1257. Found: 404.1259.

2-benzoyl-5'-methyl-3-(4-methylbenzoyl)spiro[cyclopropane-1,3'-indolin]-2'-one (4d):



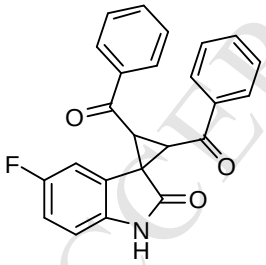
White solid, 55%. m.p. 216-218°C. IR (KBr): 3185, 3028, 1712, 1672, 1603, 1490, 1448, 1411, 1331, 1301, 1204, 1178, 1024, 808, 756. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.30 (s, 1H, NH), 7.96 (d, *J* = 7.8Hz, 2H, ArH), 7.71 (d, *J* = 8.4Hz, 2H, ArH), 7.53 (t, *J* = 7.2Hz, 1H, ArH), 7.40 (t, *J* = 7.8Hz, 2H, ArH), 7.14 (d, *J* = 7.8Hz, 2H, ArH), 7.11 (s, 1H, ArH), 7.02 (t, *J* = 7.8Hz, 1H, ArH), 6.78 (d, *J* = 7.8Hz, 1H, ArH), 4.37 (d, *J* = 7.8Hz, 1H, CH), 4.10 (d, *J* = 7.8Hz, 1H, CH), 2.32 (s, 6H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 192.3, 190.2, 173.4, 144.5, 139.1, 136.9, 133.9, 133.8, 132.3, 129.5, 129.0, 128.7, 128.6, 128.5, 124.5, 123.2, 110.1, 41.3, 39.8, 38.8, 21.6, 21.3. HRMS (ESI) Calcd. for $C_{26}H_{21}NNaO_3$ ([M+Na]$^+$): 418.1414. Found: 418.1416.

3-(2-hydroxy-5-phenyl-2-p-tolylfuran-3(2H)-ylidene)-5-methylindolin-2-one (5d):



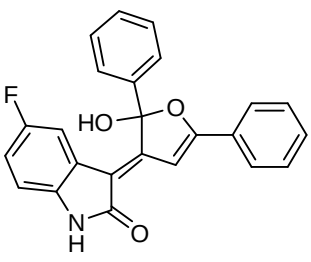
Yellow solid, 36%. m.p. 190-192°C. IR (KBr): 3424, 3206, 1702, 1647, 1616, 1484, 1448, 1351, 1261, 1212, 1147, 1114, 1008, 936, 824. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.31 (s, 1H, NH), 8.80 (s, 1H, OH), 7.93 (s, 1H, ArH), 7.78 (d, *J* = 7.2Hz, 2H, ArH), 7.51-7.47 (m, 5H, ArH), 7.21 (s, 1H, ArH), 7.17 (d, *J* = 7.8Hz, 2H, ArH), 6.75 (d, *J* = 7.8Hz, 1H, ArH), 6.57 (d, *J* = 7.8Hz, 1H, ArH), 2.26 (s, 3H, CH_3), 1.99 (s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 169.0, 166.4, 155.5, 138.3, 135.4, 131.3, 129.0, 128.9, 128.8, 128.3, 127.5, 126.4, 125.3, 125.1,121.8, 114.9, 111.3, 108.4, 100.5, 20.8, 20.7. HRMS (ESI) Calcd. for $C_{26}H_{21}NNaO_3$ ([M+Na]$^+$): 418.1414. Found: 418.1414.

(5'-fluoro-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis(phenylmethanone) (4e):



White solid, 58%. m.p. 188-190°C. IR (KBr): 3313, 3019, 1715, 1673, 1598, 1485, 1331, 1222, 1184, 1151, 1017, 945, 739. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.84 (s, 1H, NH), 7.98 (d, *J* = 7.8Hz, 2H, ArH), 7.79 (d, *J* = 7.2Hz, 2H, ArH), 7.56 (t, *J* = 7.2Hz, 1H, ArH), 7.49 (t, *J* = 7.2Hz, 1H, ArH), 7.42 (d, *J* = 7.8Hz, 2H, ArH), 7.35 (t, *J* = 7.2Hz, 2H, ArH), 7.13 (d, *J* = 7.2Hz, 1H, ArH), 6.96 (t, *J* = 7.2Hz, 1H, ArH), 6.89 (d, *J* = 7.8Hz, 1H, ArH), 4.42 (d, *J* = 7.8Hz, 1H, CH), 4.14 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.9, 190.2, 173.8, 159.1 (d, *J* = 238.8Hz), 137.9, 136.5, 136.0, 134.1, 133.8, 128.9, 128.7, 128.4, 125.8 (d, *J* = 9.2Hz), 115.4 (d, *J* = 23.2Hz), 111.6 (d, *J* = 7.9Hz), 110.7 (d, *J* = 26.4Hz), 41.5, 39.9, 38.6. HRMS (ESI) Calcd. for $C_{24}H_{16}FNNaO_3$ ([M+Na]$^+$): 408.1006. Found: 408.1010.

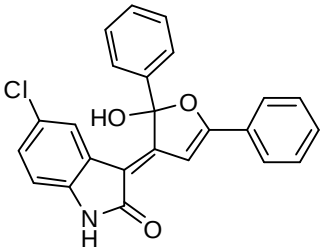
5-fluoro-3-(2-hydroxy-2,5-diphenylfuran-3(2H)-ylidene)indolin-2-one (5e):



Yellow solid, 34%. m.p. 170-172°C. IR (KBr): 3404, 1717, 1671, 1604, 1537, 1479, 1388, 1347, 1287, 1252, 1216, 1180, 1148, 936, 817. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.49 (s, 1H, NH),

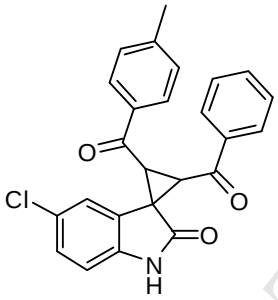
9.05 (s, 1H, OH), 7.95 (s, 1H, ArH), 7.82 (d, *J* = 7.2Hz, 2H, ArH), 7.63 (d, *J* = 6.6Hz, 2H, ArH), 7.54 (d, *J* = 6.6Hz, 1H, ArH), 7.51 (d, *J* = 7.2Hz, 2H, ArH), 7.40 (d, *J* = 7.2Hz, 3H, ArH), 7.06 (d, *J* = 9.0Hz, 1H, ArH), 6.78 (t, *J* = 7.2Hz, 1H, ArH), 6.67 (d, *J* = 7.2Hz, 1H, ArH). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.8, 167.8, 156.8 (d, *J* = 231.3Hz), 157.0, 137.7, 136.7, 131.8, 129.3, 129.1, 128.6, 126.7, 125.2, 122.7 (d, *J* = 9.3Hz), 114.2, 113.2 (d, *J* = 23.4Hz), 111.3, 110.7 (d, *J* = 26.9Hz), 109.2 (d, *J* = 8.9Hz), 100.6. HRMS (ESI) Calcd. for $C_{24}H_{16}FNNaO_3$ ([M+Na]$^+$): 408.1006. Found: 408.1006.

5-chloro-3-(2-hydroxy-2,5-diphenylfuran-3(2H)-ylidene)indolin-2-one (5f):



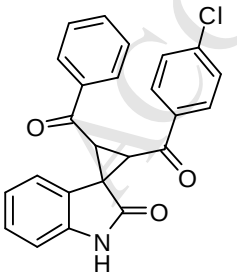
Yellow solid, 72%. m.p. 156-158°C. IR (KBr): 3449, 3404, 1657, 1539, 1480, 1417, 1386, 1349, 1320, 1249, 1201, 1144, 933, 814. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.60 (s, 1H, NH), 9.07 (s, 1H, OH), 7.94 (s, 1H, ArH), 7.82 (d, *J* = 7.2Hz, 2H, ArH), 7.62 (d, *J* = 6.6Hz, 2H, ArH), 7.54 (d, *J* = 6.6Hz, 1H, ArH), 7.50 (t, *J* = 7.2Hz, 2H, ArH), 7.39 (d, *J* = 6.6Hz, 3H, ArH), 7.31 (s, 1H, ArH), 6.98 (d, *J* = 7.8Hz, 1H, ArH), 6.70 (d, *J* = 7.8Hz, 1H, ArH). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.6, 168.1, 157.4, 139.0, 137.7, 131.8, 129.3, 129.1, 128.6, 128.5, 126.7, 126.4, 125.3, 124.2, 123.6, 123.3, 113.6, 111.4, 110.0, 100.7. HRMS (ESI) Calcd. for $C_{24}H_{16}ClNNaO_3$ ([M+Na]$^+$): 424.0711. Found: 424.0710.

2-benzoyl-5'-chloro-3-(4-methylbenzoyl)spiro[cyclopropane-1,3'-indolin]-2'-one (4g):



White solid, 62%. m.p. 222-224°C. IR (KBr): 3179, 3063, 3022, 2862, 1723, 1667, 1606, 1477, 1445, 1327, 1221, 1088, 1023, 937, 809. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.56 (s, 1H, NH), 7.99 (d, *J* = 7.8Hz, 2H, ArH), 7.69 (d, *J* = 7.8Hz, 2H, ArH), 7.56 (t, *J* = 7.2Hz, 1H, ArH), 7.43 (t, *J* = 7.2Hz, 2H, ArH), 7.36 (t, *J* = 6.6Hz, 1H, ArH), 7.22 (d, *J* = 7.8Hz, 1H, ArH), 7.15 (d, *J* = 7.8Hz, 2H, ArH), 6.86 (d, *J* = 8.4Hz, 1H, ArH), 4.40 (d, *J* = 7.8Hz, 1H, CH), 4.12 (d, *J* = 7.8Hz, 1H, CH), 2.33 (s, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.8, 189.7, 173.4, 144.9, 140.2, 136.6, 134.0, 133.5, 129.6, 129.5, 128.8, 128.8, 128.5, 128.4, 128.2, 126.1, 123.1, 111.7, 41.1, 40.1, 38.8, 21.7. HRMS (ESI) Calcd. for $C_{25}H_{18}ClNNaO_3$ ([M+Na]$^+$): 438.0867. Found: 438.0870.

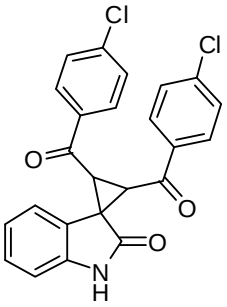
2-benzoyl-3-(4-chlorobenzoyl)spiro[cyclopropane-1,3'-indolin]-2'-one (4h):



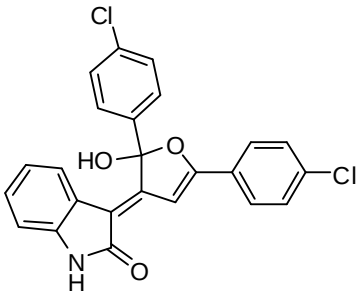
White solid, 70%. m.p. 208-210°C. IR (KBr): 3283, 1725, 1670, 1621, 1598, 1469, 1398, 1318, 1221, 1092, 1014, 934, 842, 740. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.65 (s, 1H, NH), 7.98 (d, *J* = 7.8Hz, 2H, ArH), 7.81 (d, *J* = 7.8Hz, 2H, ArH), 7.49 (t, *J* = 7.2Hz, 1H, ArH), 7.36-7.34 (m, 4H, ArH), 7.29 (d, *J* = 7.8Hz, 1H, ArH), 7.25 (d, *J* = 7.2Hz, 1H, ArH), 7.03 (t, *J* = 7.8Hz, 1H, ArH), 6.94 (d, *J* = 7.8Hz, 1H, ArH), 4.35 (d, *J* = 8.4Hz, 1H, CH), 4.16 (d, *J* = 8.4Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.0, 190.4, 173.7, 141.8, 140.5, 136.0, 134.9, 133.8, 133.0, 129.1, 128.9, 128.8, 128.4, 124.1, 122.8, 122.4, 110.0, 41.3, 39.5, 38.5. HRMS (ESI) Calcd. for $C_{24}H_{16}ClNNaO_3$ ([M+Na]$^+$): 424.0711. Found: 424.0714.

(2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis((4-chlorophenyl)methanone) (4i)：

White solid, 42%. m.p. 202-204°C. IR (KBr): 3292, 3053, 1711, 1672, 1618, 1586, 1470, 1397, 1315, 1220, 1191, 1092, 1010, 843. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.66 (s, 1H, NH), 7.88 (d, *J* = 8.4Hz, 2H, ArH), 7.73 (d, *J* = 8.4Hz, 2H, ArH), 7.35 (d, *J* = 8.4Hz, 2H, ArH), 7.33 (d, *J* = 8.4Hz, 2H, ArH), 7.28-7.25 (m, 2H, ArH), 7.03 (t, *J* = 7.8Hz, 1H, ArH), 6.95 (d, *J* = 7.2Hz, 1H, ArH), 4.32 (d, *J* = 7.2Hz, 1H, CH), 4.11 (d, *J* = 7.2Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.8, 189.4, 173.5, 141.7, 140.6, 140.3, 134.8, 134.4, 129.9, 129.8, 129.2, 129.1, 129.0, 123.9, 122.9, 122.5, 110.9, 41.2, 39.3, 38.5. HRMS (ESI) Calcd. for $C_{24}H_{15}Cl_2NNaO_3$ ([M+Na]$^+$): 458.0321. Found: 458.0324.

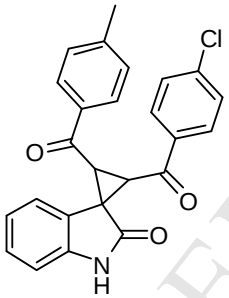


3-(2,5-bis(4-chlorophenyl)-2-hydroxyfuran-3(2H)-ylidene)indolin-2-one (5i):



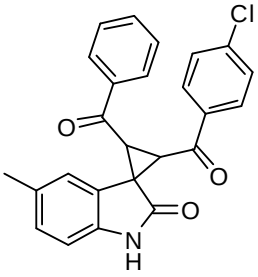
Yellow solid, 50%. m.p. 150-152°C. IR (KBr): 3438, 3168, 1658, 1611, 1571, 1542, 1485, 1406, 1352, 1266, 1177, 1152, 1096, 1053, 928, 838. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.50 (s, 1H, NH), 9.07 (s, 1H, OH), 7.96 (s, 1H, ArH), 7.81 (d, *J* = 8.4Hz, 2H, ArH), 7.62 (d, *J* = 8.4Hz, 2H, ArH), 7.55 (d, *J* = 8.4Hz, 2H, ArH), 7.44 (d, *J* = 8.4Hz, 2H, ArH), 7.27 (d, *J* = 7.2Hz, 1H, ArH), 6.98 (t, *J* = 7.2Hz, 1H, ArH), 6.71 (d, *J* = 7.8Hz, 1H, ArH), 6.60 (t, *J* = 7.8Hz, 1H, ArH). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.7, 165.0, 154.2, 140.7, 137.0, 136.0, 133.7, 129.2, 128.6, 128.2, 127.6, 127.3, 124.2, 121.4, 120.2, 115.6, 115.5, 110.8, 109.0, 101.2. HRMS (ESI) Calcd. for $C_{24}H_{15}Cl_2NNaO_3$ ([M+Na]$^+$): 458.0321. Found: 458.0320.

2-(4-chlorobenzoyl)-3-(4-methylbenzoyl)2-(4-chlorobenzoyl)-3-(4-methyl-benzoyl)spiro[cyclo propane-1,3'-indolin]-2'-one (4j):



White solid, 72%. m.p. 198-200°C. IR (KBr): 3293, 3051, 1734, 1708, 1668, 1609, 1469, 1365, 1320, 1221, 1177, 1093, 1034, 1012, 841, 749. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): *E*-isomer: 8.62 (s, 1H, NH), 7.88 (d, *J* = 7.8Hz, 2H, ArH), 7.70 (d, *J* = 7.8Hz, 2H, ArH), 7.34 (d, *J* = 8.4Hz, 2H, ArH), 7.29 (d, *J* = 7.2Hz, 1H, ArH), 7.24 (d, *J* = 7.8Hz, 1H, ArH), 7.14 (d, *J* = 7.8Hz, 2H, ArH), 7.03 (t, *J* = 7.8Hz, 1H, ArH), 6.94 (d, *J* = 7.8Hz, 1H, ArH), 4.33 (d, *J* = 7.8Hz, 1H, CH), 4.14 (d, *J* = 7.8Hz, 1H, CH), 2.32 (s, 3H, CH_3); *Z*-isomer: 8.56 (s, 1H, NH), 7.85 (d, *J* = 7.8Hz, 2H, ArH), 7.75 (d, *J* = 7.8Hz, 2H, ArH), 7.32 (d, *J* = 8.4Hz, 2H, ArH), 7.17 (d, *J* = 7.8Hz, 1H, ArH), 4.36 (d, *J* = 7.8Hz, 1H, CH), 4.12 (d, *J* = 7.8Hz, 1H, CH), 2.35 (s, 3H, CH_3); *Z*/*E* = 2:5; ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.3, 191.0, 189.9, 189.6, 173.5, 145.0, 144.5, 141.6, 140.4, 140.2, 135.0, 134.6, 133.7, 129.9, 129.8, 129.5, 129.1, 128.7, 128.5, 124.2, 122.8, 122.5, 110.7, 41.3, 41.0, 39.7, 39.3, 38.7, 38.2, 31.7. HRMS (ESI) Calcd. for $C_{25}H_{18}ClNNaO_3$ ([M+Na]$^+$): 438.0867. Found: 438.0870.

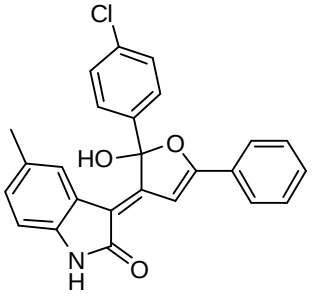
2-benzoyl-3-(4-chlorobenzoyl)-5'-methylspiro[cyclopropane-1,3'-indolin]-2'-one (4k):



White solid, 60%. m.p. 220-222°C. IR (KBr): 3187, 3032, 1714, 1670, 1626, 1592, 1489, 1447, 1329, 1301, 1205, 1092, 1020, 807. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.30 (s, 1H, NH), 7.96 (d, *J* = 7.8Hz, 2H, ArH), 7.74 (d, *J* = 8.4Hz, 2H, ArH), 7.54 (t, *J* = 7.2Hz, 1H, ArH), 7.40 (t, *J* = 7.2Hz, 2H, ArH), 7.32 (d, *J* = 8.4Hz, 2H,

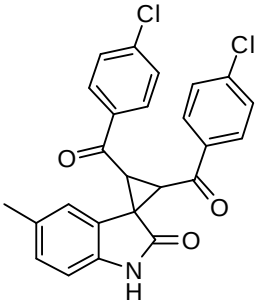
ArH), 7.10 (s, 1H, ArH), 7.04 (t, *J* = 7.2Hz, 1H, ArH), 6.80 (d, *J* = 7.8Hz, 1H, ArH), 4.36 (d, *J* = 7.8Hz, 1H, CH), 4.07 (d, *J* = 7.8Hz, 1H, CH), 2.31 (s, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 192.0, 189.6, 173.1, 140.2, 139.0, 136.7, 134.5, 134.0, 132.5, 129.8, 129.2, 128.8, 128.6, 124.1, 123.2, 110.1, 41.2, 39.4, 38.7, 21.3. HRMS (ESI) Calcd. for $C_{25}H_{18}ClNNaO_3$ ($[M+Na]^+$): 438.0867. Found: 438.0869.

3-(2-(4-chlorophenyl)-2-hydroxy-5-phenylfuran-3(2H)-ylidene)-5-methylindol-in-2-one (5k):



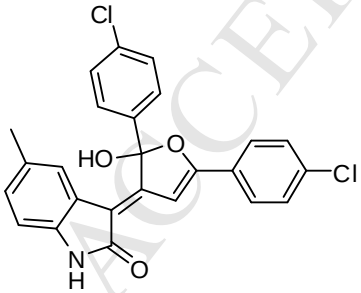
Yellow solid, 26%. m.p. 196-198°C. IR (KBr): 3444, 3163, 1655, 1544, 1484, 1406, 1337, 1268, 1209, 1088, 1006, 936, 811. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.34 (s, 1H, NH), 8.89 (s, 1H, OH), 7.96 (s, 1H, ArH), 7.81 (d, *J* = 8.4Hz, 2H, ArH), 7.63 (d, *J* = 6.6Hz, 2H, ArH), 7.54 (d, *J* = 8.4Hz, 2H, ArH), 7.37 (d, *J* = 7.8Hz, 3H, ArH), 7.13 (s, 1H, ArH), 6.76 (d, *J* = 7.8Hz, 1H, ArH), 6.57 (d, *J* = 7.8Hz, 1H, ArH), 1.96(s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.9, 165.0, 154.8, 138.4, 138.1, 135.8, 129.1, 129.0, 128.4, 128.3, 128.2, 127.8, 127.7, 125.4, 125.1, 121.6, 115.6, 111.3, 108.5, 101.1, 20.7. HRMS (ESI) Calcd. for $C_{25}H_{18}ClNNaO_3$ ($[M+Na]^+$): 438.0867. Found: 438.0866.

(5'-methyl-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis((4-chlorophenyl)methanone) (4l):



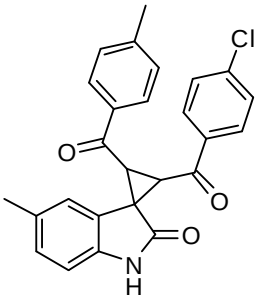
White solid, 43%. m.p. 182-184°C. IR (KBr): 3442, 1709, 1670, 1590, 1490, 1403, 1326, 1202, 1092, 1042, 1012, 822, 763. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.25 (s, 1H, NH), 7.88 (d, *J* = 8.4Hz, 2H, ArH), 7.74 (d, *J* = 8.4Hz, 2H, ArH), 7.35-7.33 (m, 4H, ArH), 7.06 (s, 1H, ArH), 7.05 (d, *J* = 7.8Hz, 1H, ArH), 6.81 (d, *J* = 7.8Hz, 1H, ArH), 4.30 (d, *J* = 7.8Hz, 1H, CH), 4.07 (d, *J* = 7.8Hz, 1H, CH), 2.32 (s, 3H, CH_3). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 192.8, 189.4, 173.2, 140.5, 140.3, 139.1, 134.9, 134.4, 132.6, 129.9, 129.8, 129.4, 129.2, 129.1, 123.9, 123.1, 110.3, 41.2, 39.3, 38.5, 21.3. HRMS (ESI) Calcd. for $C_{25}H_{17}Cl_2NNaO_3$ ($[M+Na]^+$): 472.0478. Found: 472.0478.

3-(2,5-bis(4-chlorophenyl)-2-hydroxyfuran-3(2H)-ylidene)-5-methylindolin-2- one (5l):



Yellow solid, 44%. m.p. 152-154°C. IR (KBr): 3435, 3199, 1653, 1545, 1485, 1405, 1338, 1266, 1211, 1149, 1095, 1052, 1009, 938, 842. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.37 (s, 1H, NH), 9.04 (s, 1H, OH), 7.95 (s, 1H, ArH), 7.81 (d, *J* = 8.4Hz, 2H, ArH), 7.63 (d, *J* = 7.8Hz, 2H, ArH), 7.54 (d, *J* = 8.4Hz, 2H, ArH), 7.45 (d, *J* = 7.8Hz, 2H, ArH), 7.13 (s, 1H, ArH), 6.79 (d, *J* = 7.8Hz, 1H, ArH), 6.69 (d, *J* = 7.8Hz, 1H, ArH), 1.99 (s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.8, 164.9, 154.1, 138.5, 137.1, 135.9, 133.7, 129.2, 128.5, 128.4, 128.2, 128.0, 127.6, 127.3, 125.0, 121.4, 115.8, 110.6, 108.6, 101.1, 20.8. HRMS (ESI) Calcd. for $C_{25}H_{17}Cl_2NNaO_3$ ($[M+Na]^+$): 472.0478. Found: 472.0474.

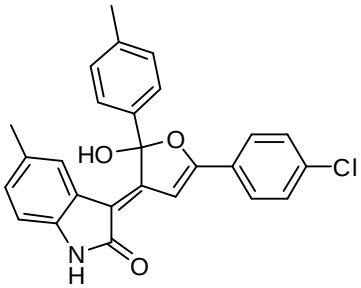
2-(4-chlorobenzoyl)-5'-methyl-3-(4-methylbenzoyl)spiro[cyclop ropane-1,3'-indolin]-2'-one (4m):



White solid, 32%. m.p. 212-214°C. IR (KBr): 3297, 3023, 2917,

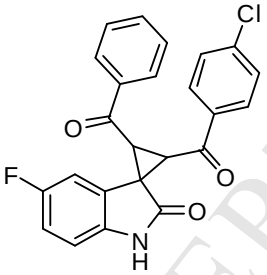
1717, 1674, 1597, 1489, 1400, 1325, 1184, 1093, 1039, 822. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): *E*-isomer: 8.20 (s, 1H, NH), 7.89 (d, *J* = 8.4Hz, 2H, ArH), 7.70 (d, *J* = 7.8Hz, 2H, ArH), 7.34 (d, *J* = 8.4Hz, 2H, ArH), 7.16 (d, *J* = 7.8Hz, 2H, ArH), 7.08 (s, 1H, ArH), 7.04 (d, *J* = 7.8Hz, 1H, ArH), 6.80 (d, *J* = 7.2Hz, 1H, ArH), 4.31 (d, *J* = 7.8Hz, 1H, CH), 4.09 (d, *J* = 7.8Hz, 1H, CH), 2.34 (s, 3H, CH_3), 2.32 (s, 3H, CH_3). *Z*-isomer: 8.16 (s, 1H, NH), 7.85 (d, *J* = 7.8Hz, 2H, ArH), 7.75 (d, *J* = 7.8Hz, 2H, ArH), 7.18 (d, *J* = 8.4Hz, 2H, ArH), 4.34 (d, *J* = 7.8Hz, 1H, CH), 4.07 (d, *J* = 7.8Hz, 1H, CH), 2.36 (s, 3H, CH_3), 2.31 (s, 3H, CH_3); *Z*/*E* = 1:4. ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 191.5, 191.1, 190.2, 189.8, 173.7, 144.9, 144.7, 140.3, 140.1, 139.5, 135.0, 134.6, 134.2, 133.7, 132.3, 129.9, 129.8, 129.5, 129.3, 129.1, 128.7, 128.6, 124.2, 122.9, 110.7, 41.4, 41.2, 39.5, 39.2, 38.7, 38.6, 21.7, 21.3. HRMS (ESI) Calcd. for $C_{26}H_{20}ClNNaO_3$ ([M+Na]$^+$): 452.1024. Found: 452.1025.

3-(5-(4-chlorophenyl)-2-hydroxy-2-p-tolylfuran-3(2H)-ylidene)-5-methylindolin-2-one (5m):



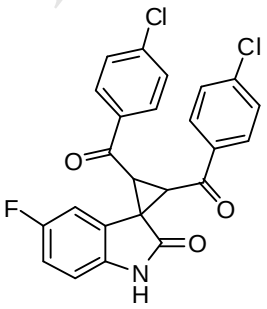
Yellow solid, 47%. m.p. 188-190°C. IR (KBr): 3440, 3220, 1654, 1571, 1543, 1482, 1406, 1383, 1334, 1266, 1220, 1150, 1111, 1090, 935, 811. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.33 (s, 1H, NH), 8.83 (s, 1H, OH), 7.94 (s, 1H, ArH), 7.80 (d, *J* = 8.4Hz, 2H, ArH), 7.54 (d, *J* = 7.8Hz, 2H, ArH), 7.50 (d, *J* = 7.8Hz, 2H, ArH), 7.19 (d, *J* = 8.4Hz, 2H, ArH), 7.16 (s, 1H, ArH), 6.76 (d, *J* = 7.2Hz, 1H, ArH), 6.57 (d, *J* = 7.8Hz, 1H, ArH), 2.26 (s, 3H, CH_3), 1.98 (s, 3H, CH_3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.9, 165.0, 155.0, 138.3, 135.7, 135.3, 129.1, 128.8, 128.4, 128.1, 127.9, 127.8, 127.7, 125.3, 121.7, 115.5, 111.4, 108.4, 105.4, 101.0, 20.8, 20.7. HRMS (ESI) Calcd. for $C_{26}H_{20}ClNNaO_3$ ([M+Na]$^+$): 452.1024. Found: 452.1022.

2-benzoyl-3-(4-chlorobenzoyl)-5'-fluorospiro[cyclopropane-1,3'-indolin]-2'-one (4n):



White solid, 76%. m.p. 204-206°C. IR (KBr): 3444, 3013, 1725, 1696, 1665, 1589, 1484, 1401, 1329, 1229, 1183, 1150, 1091, 1036, 949, 850. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.63 (s, 1H, NH), 7.92 (d, *J* = 8.4Hz, 2H, ArH), 7.79 (d, *J* = 7.8Hz, 2H, ArH), 7.51 (t, *J* = 7.2Hz, 1H, ArH), 7.40 (t, *J* = 8.4Hz, 2H, ArH), 7.37 (d, *J* = 7.2Hz, 2H, ArH), 7.11 (d, *J* = 7.8Hz, 1H, ArH), 6.98 (t, *J* = 8.4Hz, 1H, ArH), 6.89 (d, *J* = 7.8Hz, 1H, ArH), 4.36 (d, *J* = 7.8Hz, 1H, CH), 4.12 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.7, 190.0, 173.4, 159.1 (d, *J* = 239.3Hz), 140.7, 137.6, 135.9, 134.8, 133.9, 130.0, 129.2, 128.9, 12.84, 125.6 (d, *J* = 7.1Hz), 115.5 (d, *J* = 24.3Hz), 111.4 (d, *J* = 8.1Hz), 110.8 (d, *J* = 25.3Hz), 41.4, 40.0, 38.5. HRMS (ESI) Calcd. for $C_{24}H_{15}FClNNaO_3$ ([M+Na]$^+$): 442.0617. Found: 442.0619.

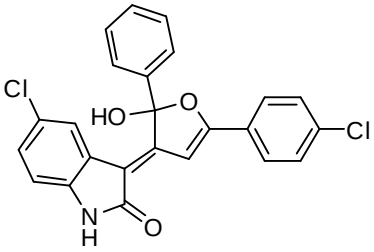
(5'-fluoro-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis((4-chlorophenyl)methanone) (4o):



White solid, 80%. m.p. 200-202°C. IR (KBr): 3251, 1721, 1677, 1587, 1480, 1400, 1324, 1224, 1179, 1092, 1011, 841, 811. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.07 (s, 1H, NH), 7.93 (d, *J* = 8.4Hz, 2H, ArH), 7.74 (d, *J* = 8.4Hz, 2H, ArH), 7.42 (t, *J* = 8.4Hz, 2H, ArH), 7.37 (t, *J* = 8.4Hz, 2H, ArH), 7.10 (d, *J* = 8.4Hz, 1H, ArH), 6.98 (t, *J* = 8.4Hz, 1H, ArH), 6.86 (d, *J* = 7.8Hz, 1H, ArH),

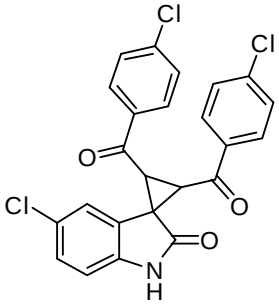
4.35 (d, *J* = 7.8Hz, 1H, CH), 4.08 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.5, 188.9, 173.0, 159.1 (d, *J* = 239.7Hz), 140.9, 140.5, 137.4, 134.7, 134.2, 130.0, 129.7, 129.5, 129.3, 125.4 (d, *J* = 7.5Hz), 115.5 (d, *J* = 23.7Hz), 111.2 (d, *J* = 8.1Hz), 110.9 (d, *J* = 26.3Hz), 41.2, 39.8, 38.5. HRMS (ESI) Calcd. for $C_{24}H_{14}FCl_2NNaO_3$ ($[M+Na]^+$): 476.0227. Found: 476.0229.

5-chloro-3-(5-(4-chlorophenyl)-2-hydroxy-2-phenylfuran-3(2H)-ylidene)indolin-2-one (5p):



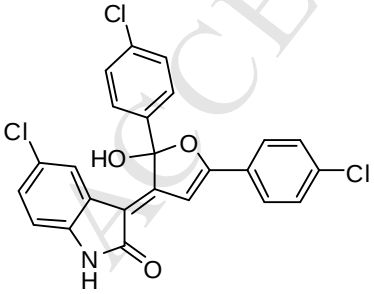
Yellow solid, 78%. m.p. 180-182°C. IR (KBr): 3440, 3178, 1660, 1591, 1542, 1489, 1449, 1408, 1340, 1239, 1170, 1112, 1081, 981, 813. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.63 (s, 1H, NH), 9.11 (s, 1H, OH), 7.96 (s, 1H, ArH), 7.85 (d, *J* = 7.2Hz, 2H, ArH), 7.62 (d, *J* = 7.2Hz, 2H, ArH), 7.56 (d, *J* = 7.2Hz, 2H, ArH), 7.39 (brs, 3H, ArH), 7.30 (s, 1H, ArH), 6.99 (d, *J* = 7.2Hz, 1H, ArH), 6.70 (d, *J* = 7.2Hz, 1H, ArH). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.5, 166.6, 156.9, 139.1, 137.6, 136.3, 129.3, 129.2, 128.5, 128.4, 127.4, 126.5, 125.3, 124.2, 123.7, 123.1, 114.1, 111.5, 110.1, 101.2. $C_{24}H_{15}Cl_2NNaO_3$ ($[M+Na]^+$): 458.0321. Found: 458.0322.

(5'-chloro-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis((4-chlorophenyl)methanone) (4q):



White solid, 49%. m.p. 210-212°C. IR (KBr): 3290, 3049, 1721, 1672, 1618, 1594, 1472, 1399, 1315, 1220, 1191, 1092, 1020, 843. ^{1}H NMR (600MHz, $CDCl_3$) δ(ppm): 8.34 (s, 1H, NH), 7.92 (d, *J* = 8.4Hz, 2H, ArH), 7.73 (d, *J* = 8.4Hz, 2H, ArH), 7.42 (d, *J* = 8.4Hz, 2H, ArH), 7.36 (d, *J* = 8.4Hz, 2H, ArH), 7.32 (d, *J* = 7.8Hz, 1H, ArH), 7.25 (d, *J* = 7.8Hz, 1H, ArH), 6.88 (d, *J* = 7.8Hz, 1H, ArH), 4.33 (d, *J* = 7.8Hz, 1H, CH), 4.09 (d, *J* = 7.8Hz, 1H, CH). ^{13}C NMR (150MHz, $CDCl_3$) δ(ppm): 190.5, 188.8, 172.9, 140.9, 140.6, 140.0, 134.7, 134.2, 130.1, 129.7, 129.3, 129.2, 129.0, 128.5, 125.6, 123.1, 111.7, 40.9, 39.8, 38.5. HRMS (ESI) Calcd. for $C_{24}H_{14}Cl_3NNaO_3$ ($[M+Na]^+$): 491.9931. Found: 491.9932.

3-(2,5-bis(4-chlorophenyl)-2-hydroxyfuran-3(2H)-ylidene)-5-chloroindolin-2-one (5q):

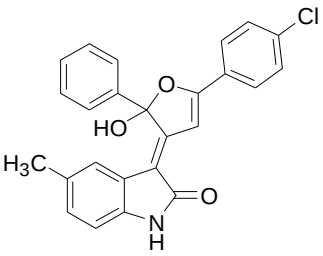


Yellow solid, 32%. m.p. 184-186°C. IR (KBr): 3399, 3194, 1659, 1611, 1537, 1481, 1446, 1348, 1250, 1203, 1169, 1110, 1075, 975, 815. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.64 (s, 1H, NH), 9.24 (s, 1H, OH), 7.95 (s, 1H, ArH), 7.84 (d, *J* = 7.8Hz, 2H, ArH), 7.62 (d, *J* = 7.8Hz, 2H, ArH), 7.56 (d, *J* = 8.4Hz, 2H, ArH), 7.47 (d, *J* = 8.4Hz, 2H, ArH), 7.27 (s, 1H, ArH), 7.00 (s, 1H, ArH), 6.72 (d, *J* = 7.8Hz, 1H, ArH). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.7, 164.7, 154.0, 138.4, 137.0, 135.7, 133.5, 129.0, 128.4, 128.3, 128.0, 127.8, 127.5, 127.2, 124.8, 121.3, 115.6, 110.4, 108.5, 101.0. HRMS (ESI) Calcd. for $C_{24}H_{14}Cl_3NNaO_3$ ($[M+Na]^+$): 491.9931. Found: 491.9926.

3. General procedure for the preparation of 3-furan-3(2H)-ylidene)indolin-2-ones from the reactions of *N*-phenacyl-4-dimethylaminopyrididinium salts with 3-phenacylideneoxindoles:

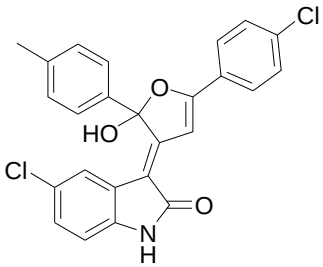
A mixture of *N*-phenacyl-4-dimethylaminopyridinium bromide or *N*-*p*-chlorophenacyl-4-dimethylaminopyridinium chloride (1.1 mmol) 3-phenacylideneoxindoles (1.0 mmol) and triethylamine (0.2 mmol) in acetonitrile (10mL) was stirred at room temperature for six hours. The resulting precipitates were collected by filtration, which was collected by filtration and washed with cold ethanol to give pure product for analysis.

3-(5-(4-chlorophenyl)-2-hydroxy-2-phenylfuran-3(2H)-ylidene)-5-methylindolin-2-one (5r):



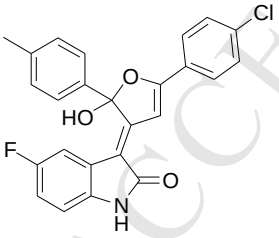
Red solid, 80%. m.p. 216-218°C. IR (KBr): 3433, 2025, 1671, 1569, 1545, 1481, 1454, 1427, 1405, 1383, 1346, 1322, 1257, 1196, 1151, 1085, 1067, 1007, 933, 880, 821, 802, 767, 733, 699, 644. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.36 (s, 1H, NH), 9.01 (s, 1H, OH), 7.99 (s, 1H, CH), 7.81 (d, *J* = 8.4Hz, 2H, ArH), 7.65 (d, *J* = 7.2Hz, 2H, ArH), 7.54 (d, *J* = 8.4Hz, 2H, ArH), 7.38 (q, *J* = 7.8Hz, 3H, ArH), 7.14 (s, 1H, ArH), 6.76 (d, *J* = 7.8Hz, 1H, ArH), 6.58 (d, *J* = 7.8Hz, 1H, ArH), 1.97 (s, 3H, CH3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.9, 165.2, 154.8, 138.3, 138.1, 135.8, 129.1, 129.0, 128.4, 128.3, 128.2, 127.6, 125.4, 125.0, 121.7, 115.3, 108.4, 101.0,20.7. HRMS (ESI) Calcd. for $C_{25}H_{18}ClNNaO_3$ ([M+Na]$^+$): 438.0867. Found: 438.0865.

5-chloro-3-(5-(4-chlorophenyl)-2-hydroxy-2-*p*-tolylfuran-3(2H)-ylidene)indolin-2-one(5s):



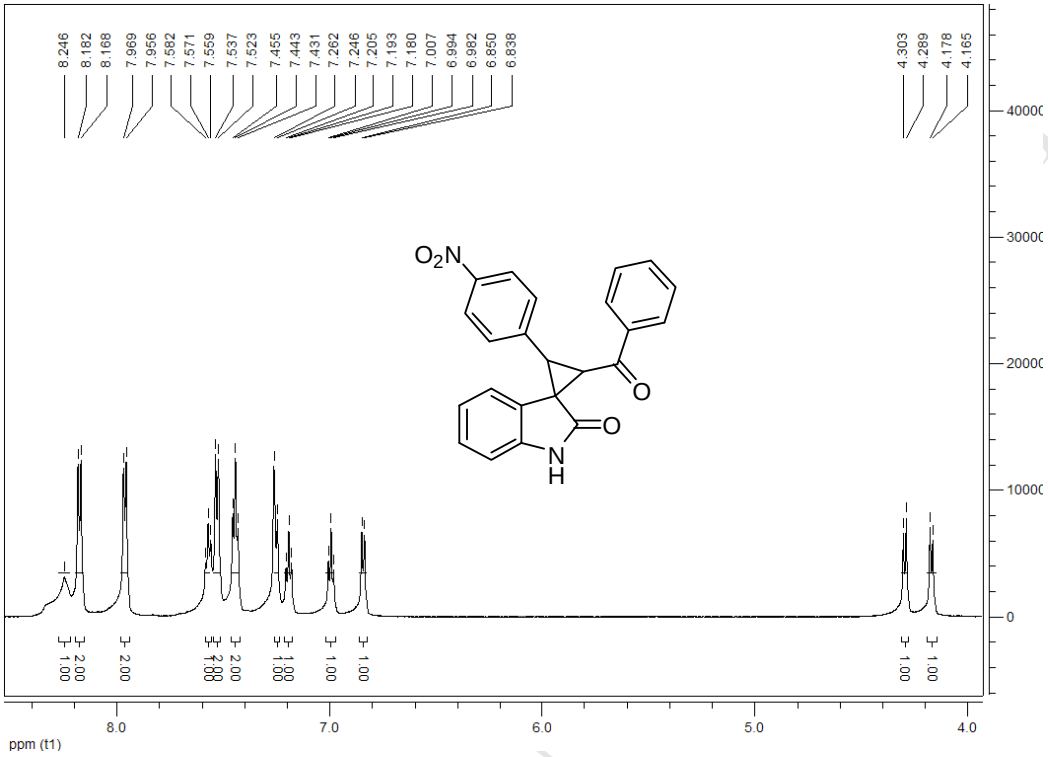
Red solid, 90%. m.p. 194-196°C. IR (KBr): 3445, 2025, 1666, 1570, 1543, 1485, 1445, 1407, 1347, 1222, 1183, 1110, 1091, 1055, 1010, 982, 932, 872, 835, 812, 718, 693, 652. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.62 (s, 1H, NH), 9.05 (s, 1H, OH), 7.95 (s, 1H, CH), 7.84 (d, *J* = 7.8Hz, 2H, ArH), 7.65 (d, *J* = 8.4Hz, 2H, ArH), 7.50 (d, *J* = 7.8Hz, 2H, ArH), 7.36 (s, 1H, ArH), 7.20 (d, *J* = 7.8Hz, 1H, ArH), 6.70 (d, *J* = 7.8Hz, 1H, ArH), 2.28 (s, 3H, CH3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.5, 166.6, 157.1, 139.1, 136.3, 134.7, 129.2, 129.0, 128.5, 127.5, 126.5, 125.2, 124.2, 123.8, 123.2, 113.9, 111.6, 110.0, 101.1, 20.7. HRMS (ESI) Calcd. for $C_{25}H_{17}Cl_2NNaO_3$ ([M+Na]$^+$): 472.0478. Found: 472.0485.

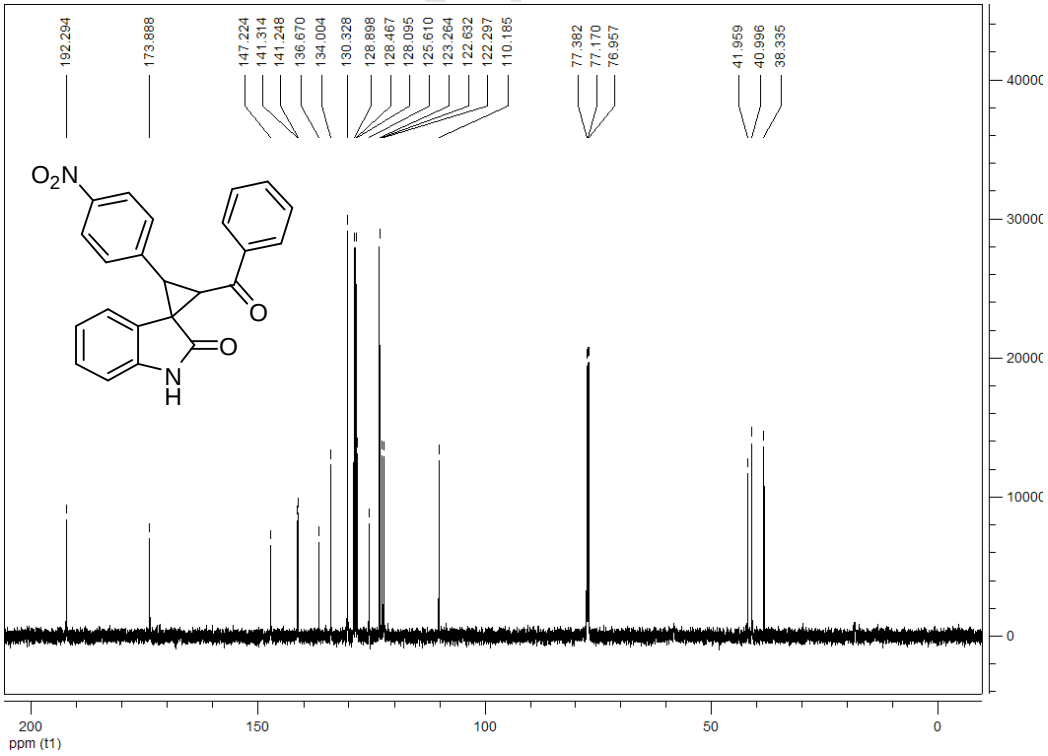
3-(5-(4-chlorophenyl)-2-hydroxy-2-p-tolylfuran-3(2H)-ylidene)-5-fluoroindolin-2-one(5t):



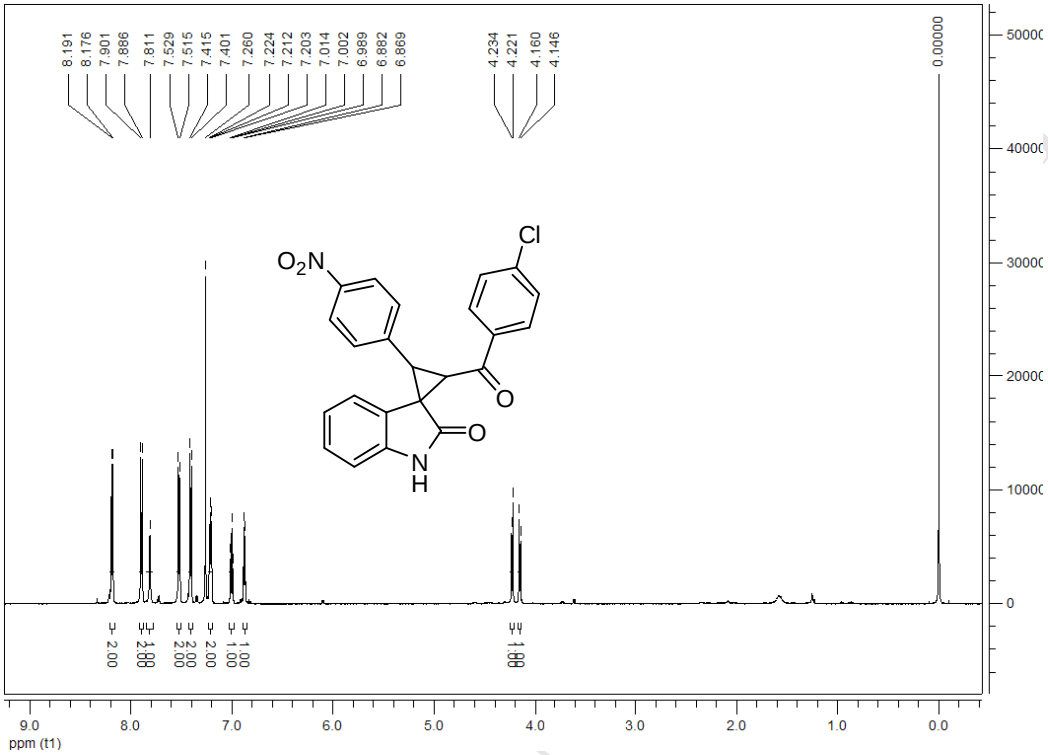
Red solid, 92%. m.p. 176-178°C. IR (KBr): 3615, 3438, 2025, 1655, 1570, 1543, 1481, 1408, 1383, 1351, 1290, 1263, 1243, 1186, 1149, 1112, 1092, 1055, 1011, 992, 938, 866, 834, 810, 698, 654, 625. ^{1}H NMR (600MHz, DMSO-d_6) δ(ppm): 10.50 (s, 1H, NH), 9.03 (s, 1H, OH), 7.95 (s, 1H, CH), 7.83 (d, *J* = 7.8Hz, 2H, ArH), 7.55 (d, *J* = 8.4Hz, 2H, ArH), 7.50 (d, *J* = 7.8Hz, 2H, ArH), 7.20 (d, *J* = 7.8Hz, 2H, ArH), 7.09 (d, *J* = 9.6Hz, 1H, ArH), 6.80 (t, *J* = 8.4Hz, 1H, ArH), 6.68(mix, 1H, ArH), 2.28 (s, 3H, CH3). ^{13}C NMR (150MHz, DMSO-d_6) δ(ppm): 168.8, 166.3, 157.2(d, *J* = 119.7Hz), 138.7, 136.8, 136.2, 134.8, 129.2, 129.1, 128.4, 127.5, 125.2, 122.7(d, *J* = 9.0Hz), 114.7(d, *J* = 11.6Hz), 113.3(d, *J* = 24.0Hz), 111.6, 110.9, 109.2(d, *J* = 7.5Hz), 101.1, 20.7. HRMS (ESI) Calcd. for $C_{25}H_{17}ClFNNaO_3$ ([M+Na]$^+$): 456.0773. Found: 456.0775.

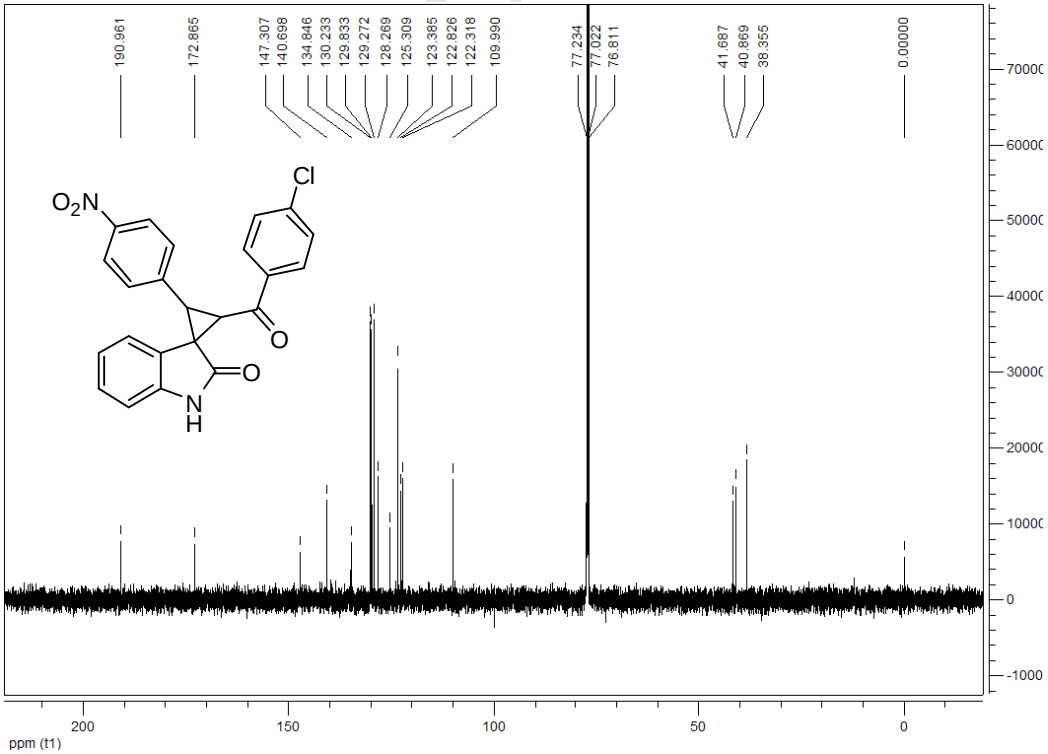
2-benzoyl-3-(4-nitrophenyl)spiro[cyclopropane-1,3'-indolin]-2'-one (3a):



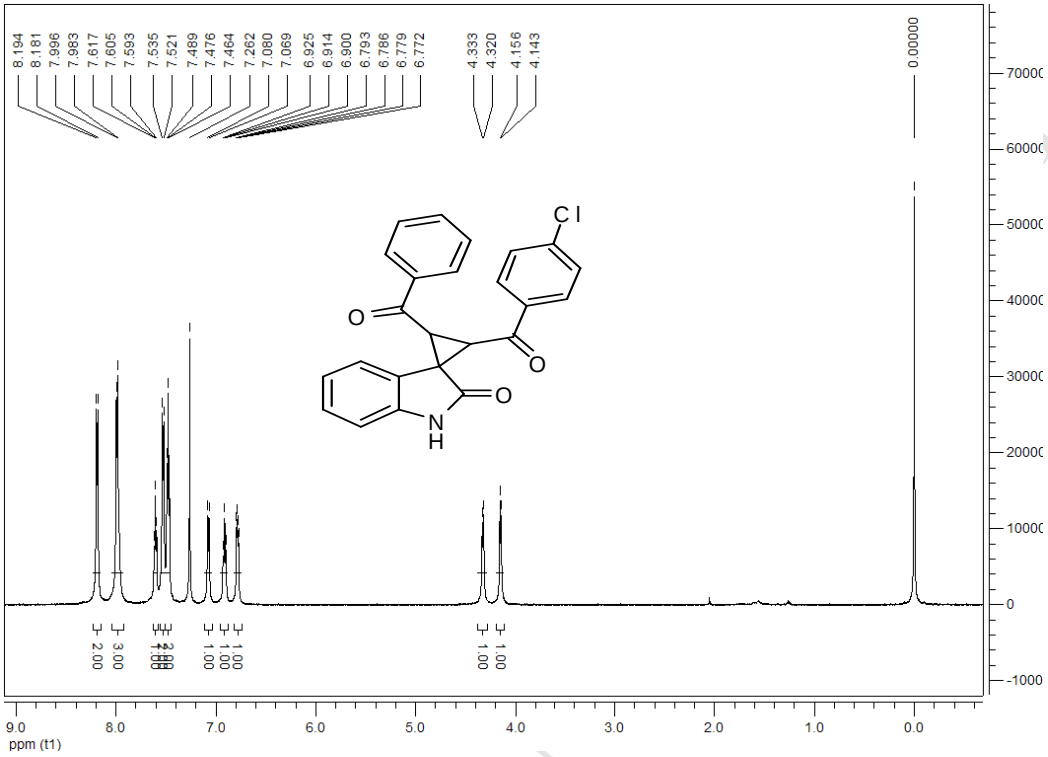


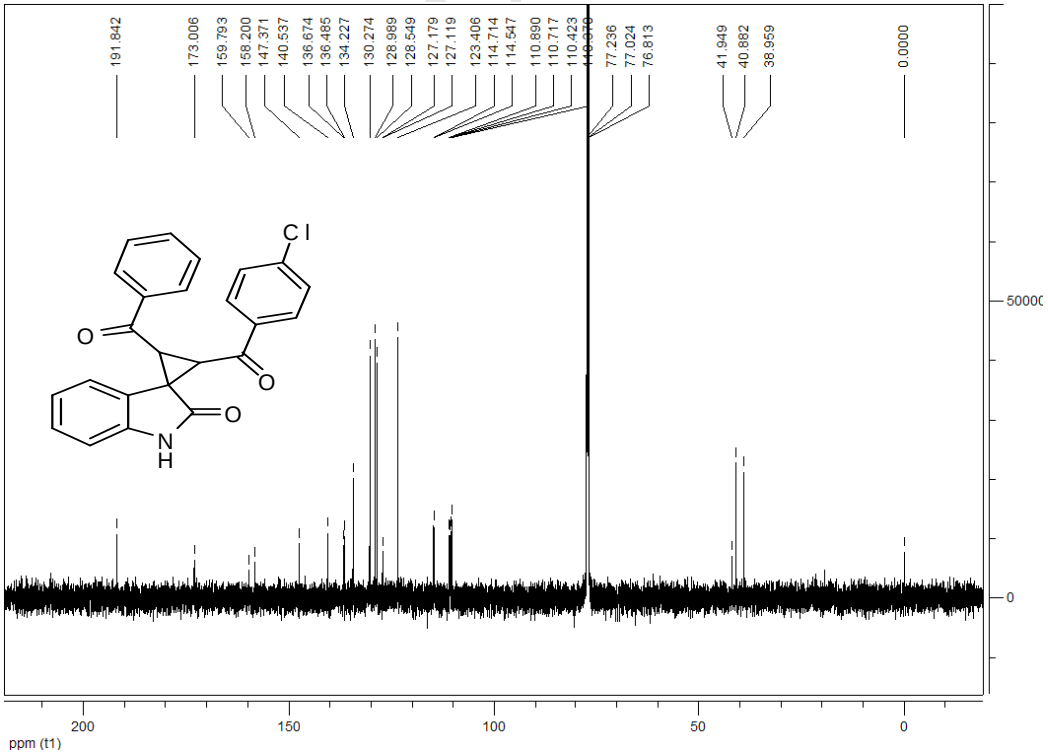
2-(4-chlorobenzoyl)-3-(4-nitrophenyl)spiro[cyclopropane-1,3'-indolin]-2'-one (3b):



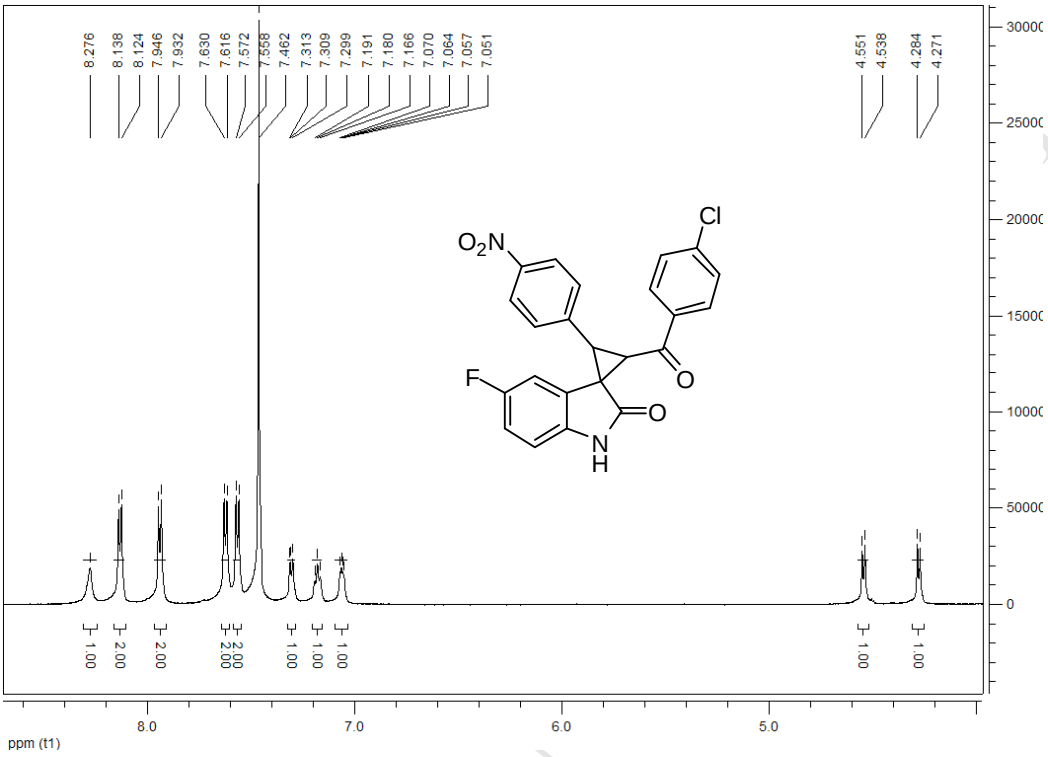


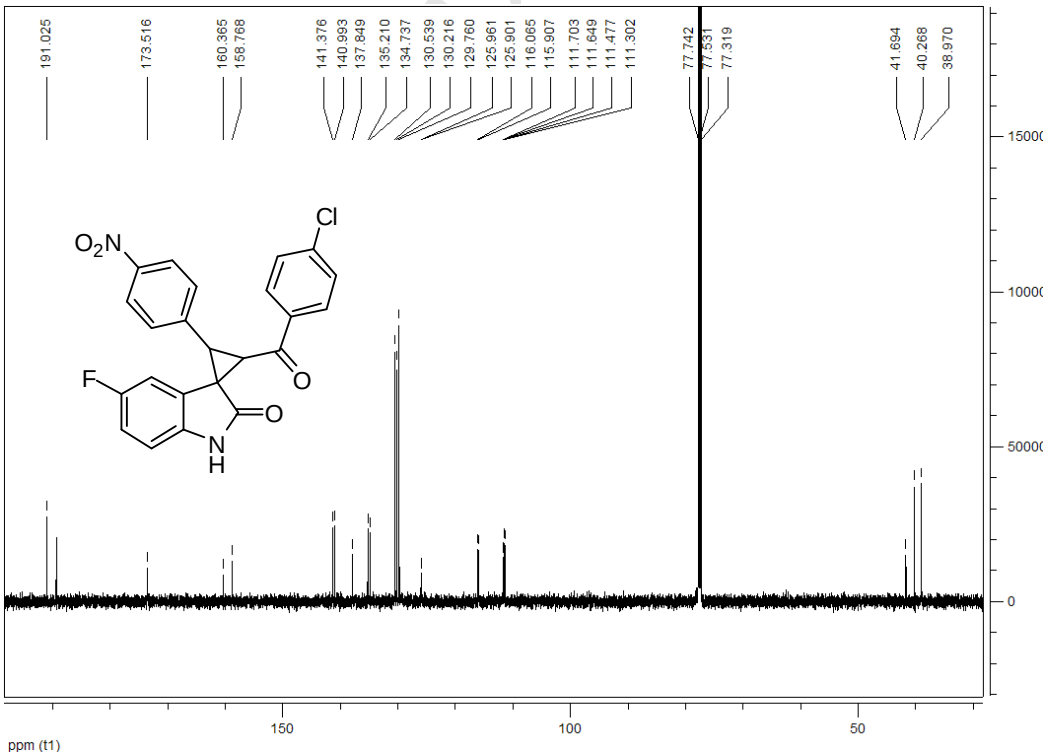
2-benzoyl-5'-fluoro-3-(4-nitrophenyl)spiro[cyclopropane-1,3'-indolin]-2'-one (3c):



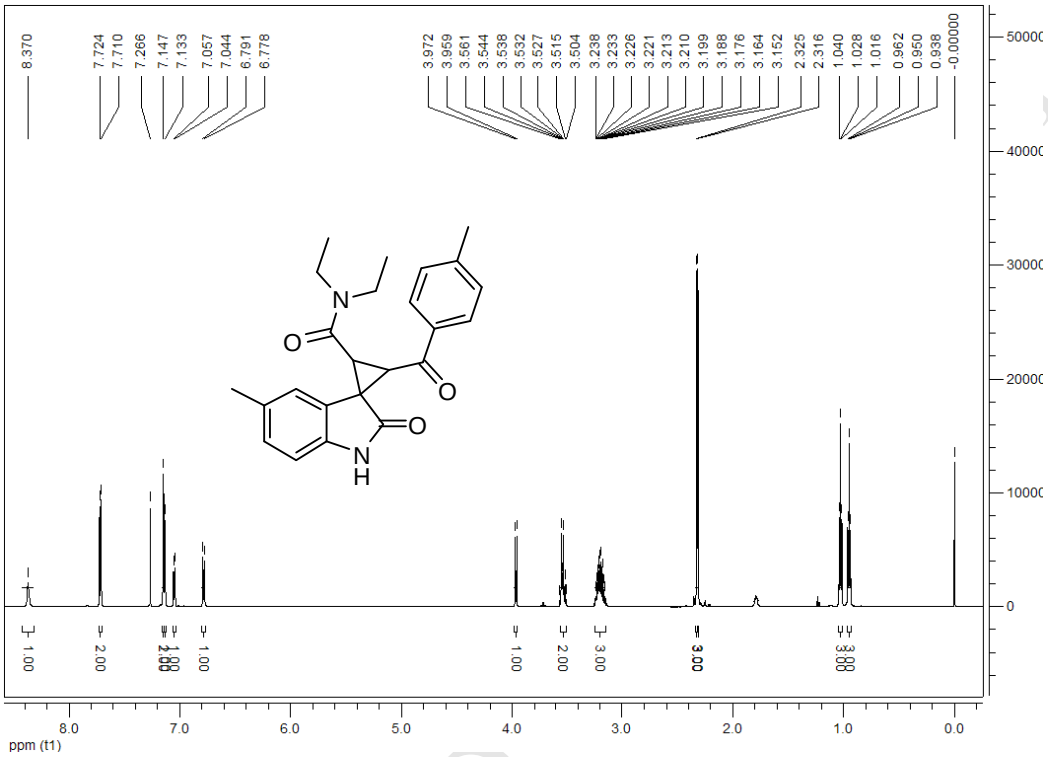


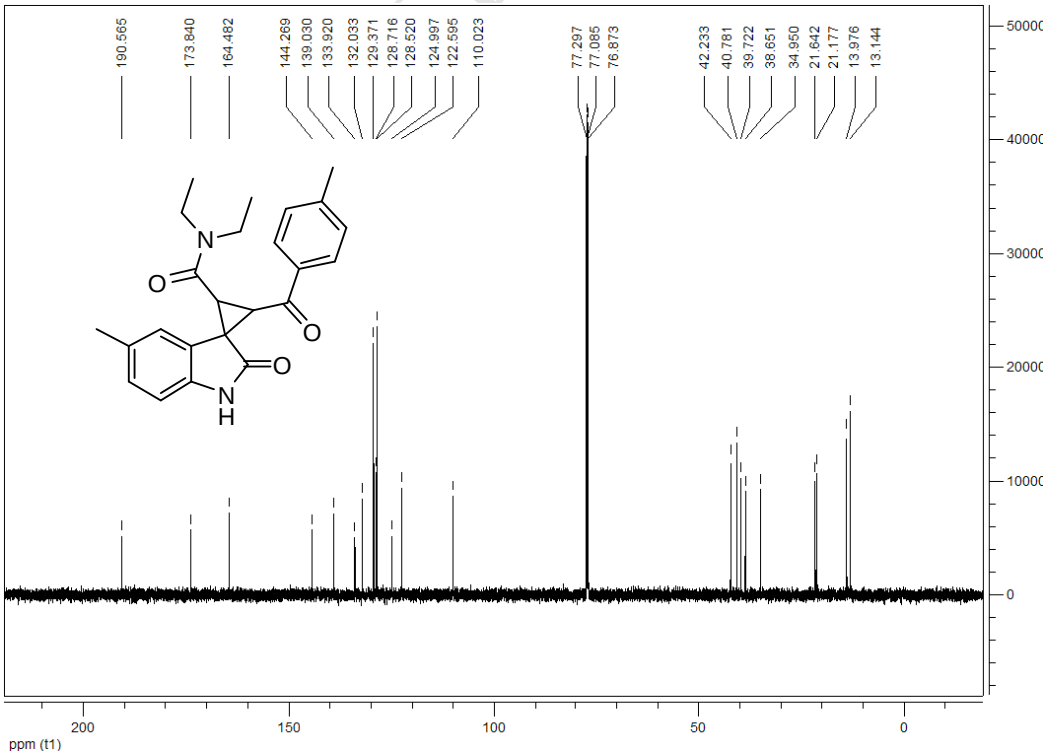
2-(4-chlorobenzoyl)-5'-fluoro-3-(4-nitrophenyl)spiro[cyclopropane-1,3'-indolin]-2'-one (3d):



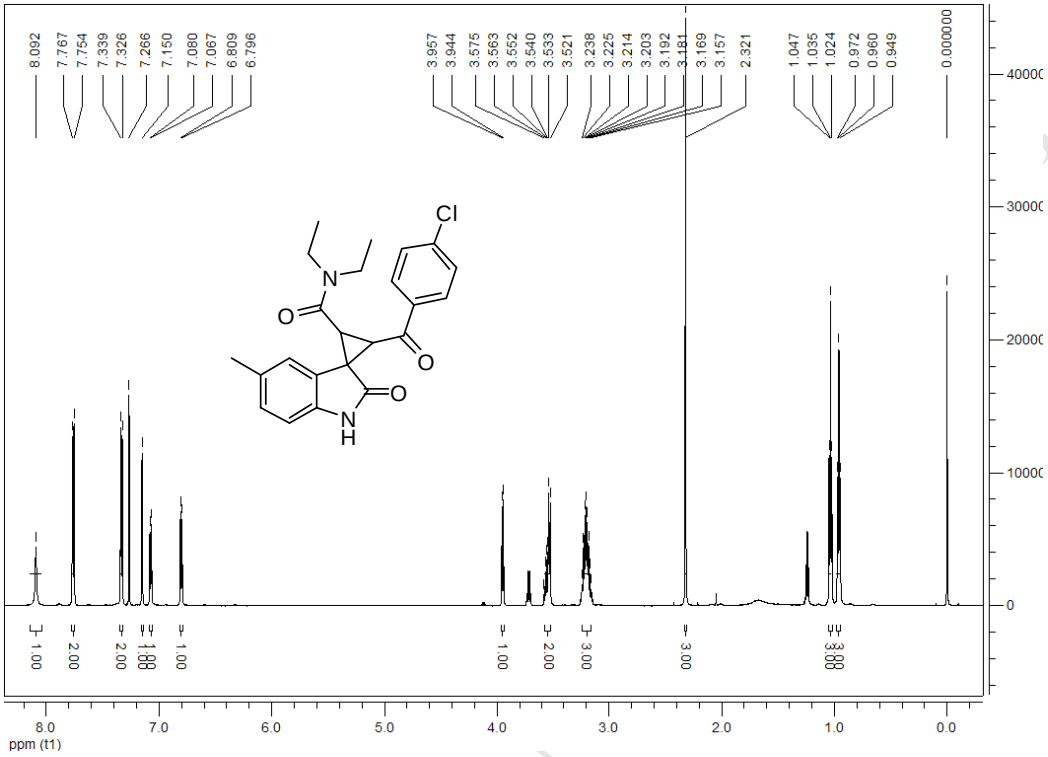


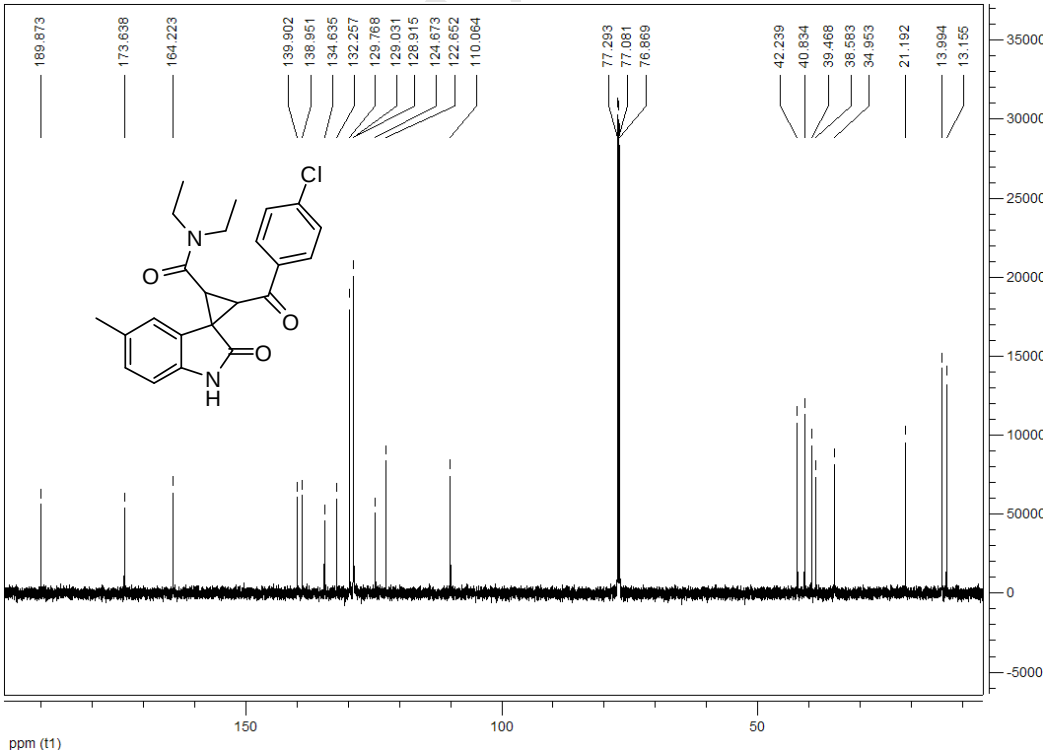
N,N-diethyl-5'-methyl-3-(4-methylbenzoyl)-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carbox amide (3e):



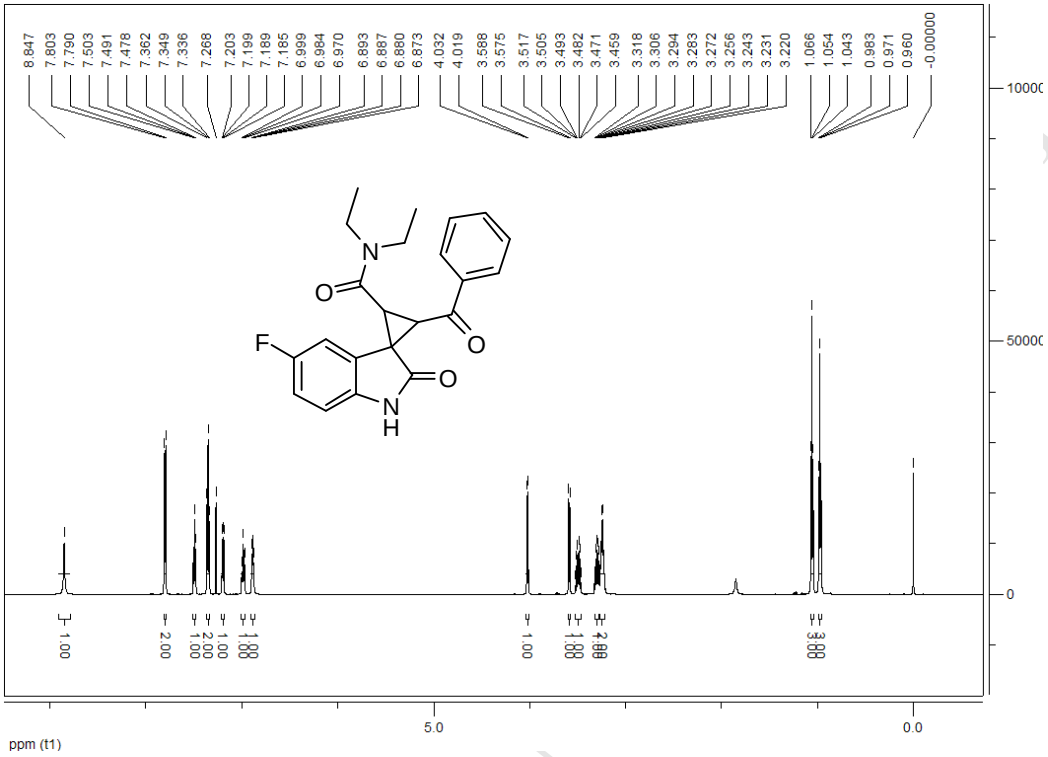


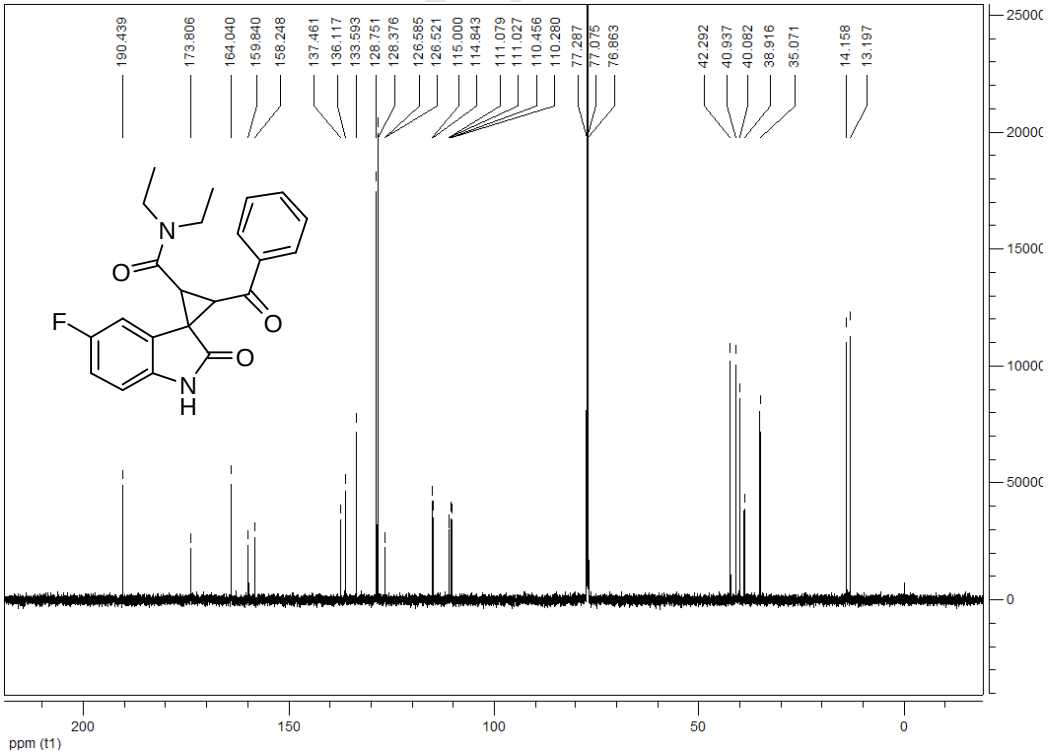
3-(4-chlorobenzoyl)-N,N-diethyl-5'-methyl-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carbox amide (3f):



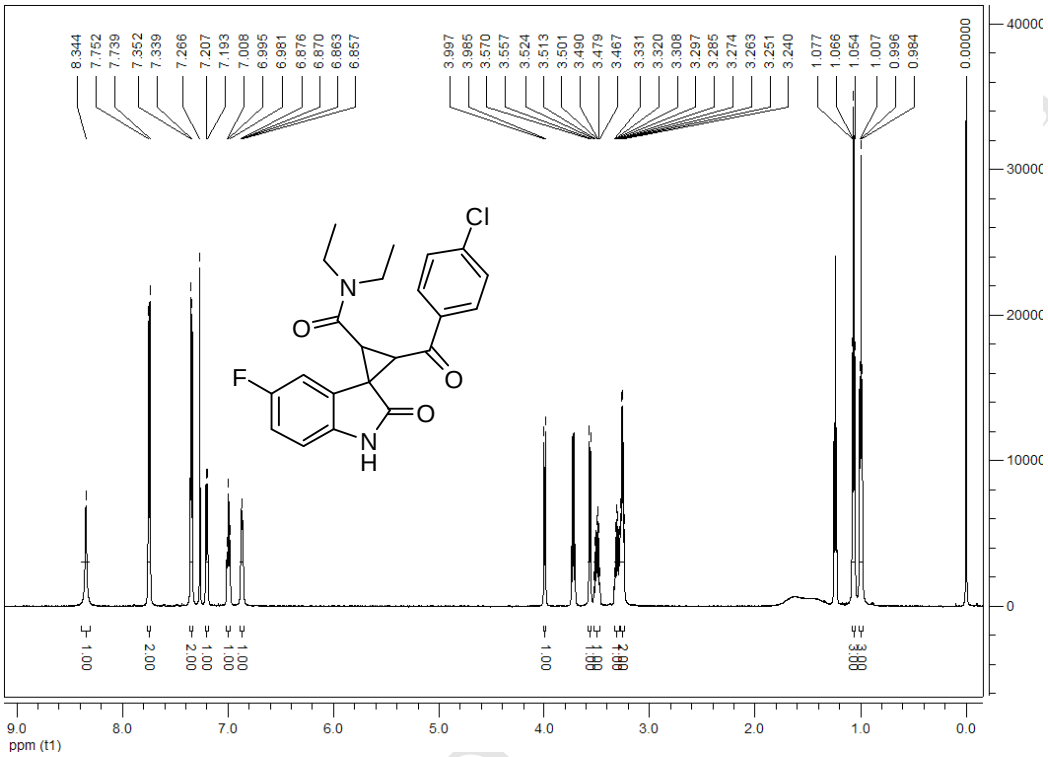


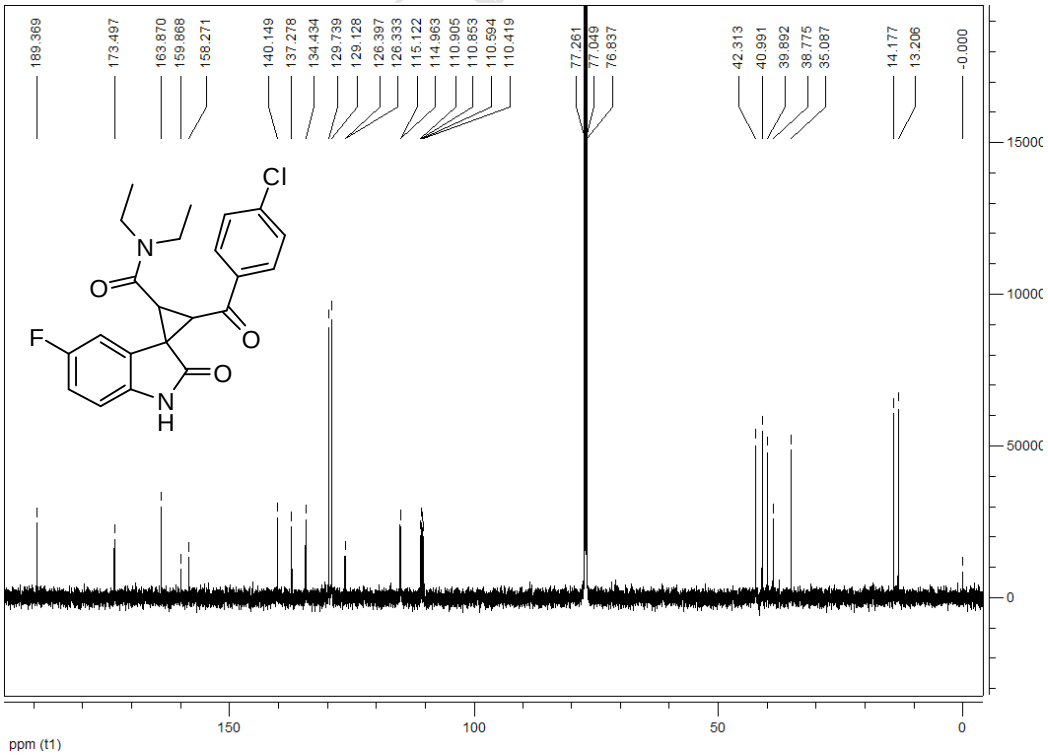
2-benzoyl-N,N-diethyl-5'-fluoro-2'-oxospiro[cyclopropane-1,3'-indoline]-3-carboxamide (3g):



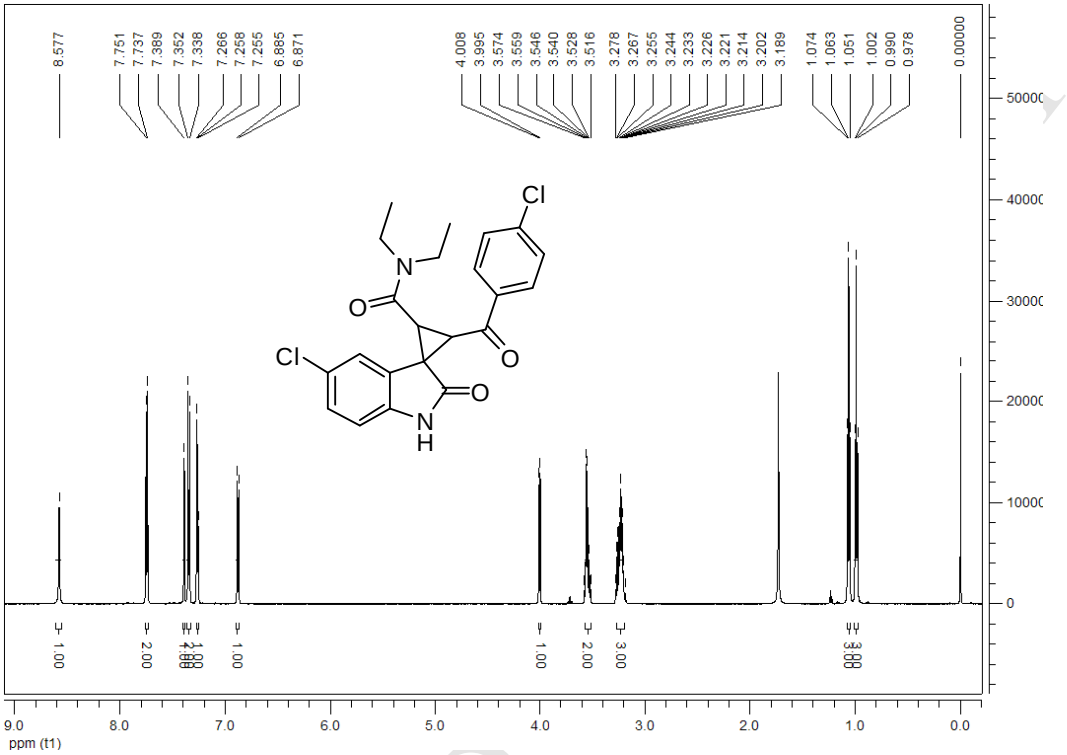


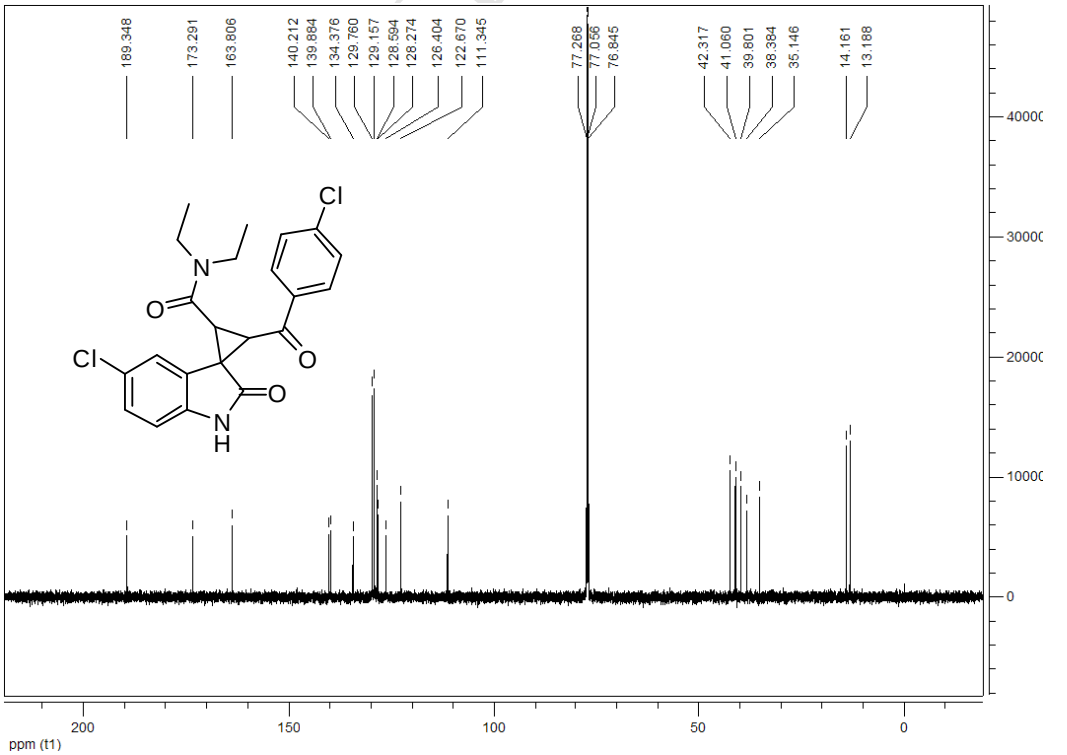
3-(4-chlorobenzoyl)-N,N-diethyl-5'-fluoro-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carboxamide (3h):



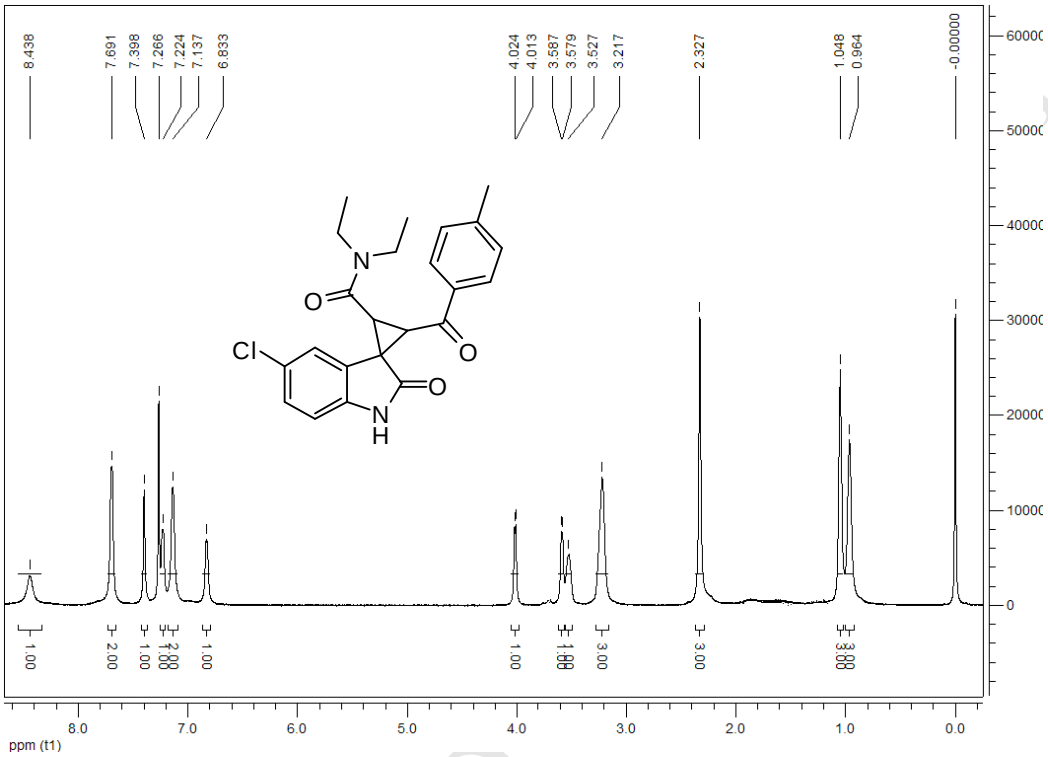


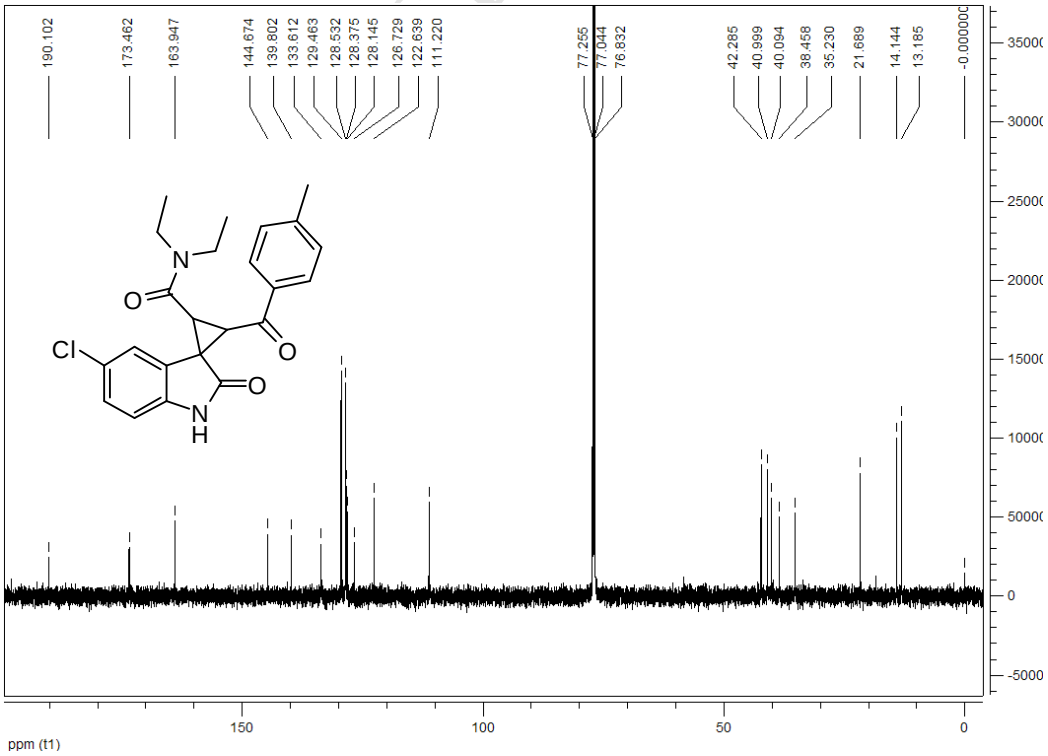
5'-chloro-3-(4-chlorobenzoyl)-N,N-diethyl-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carboxamide (3i):



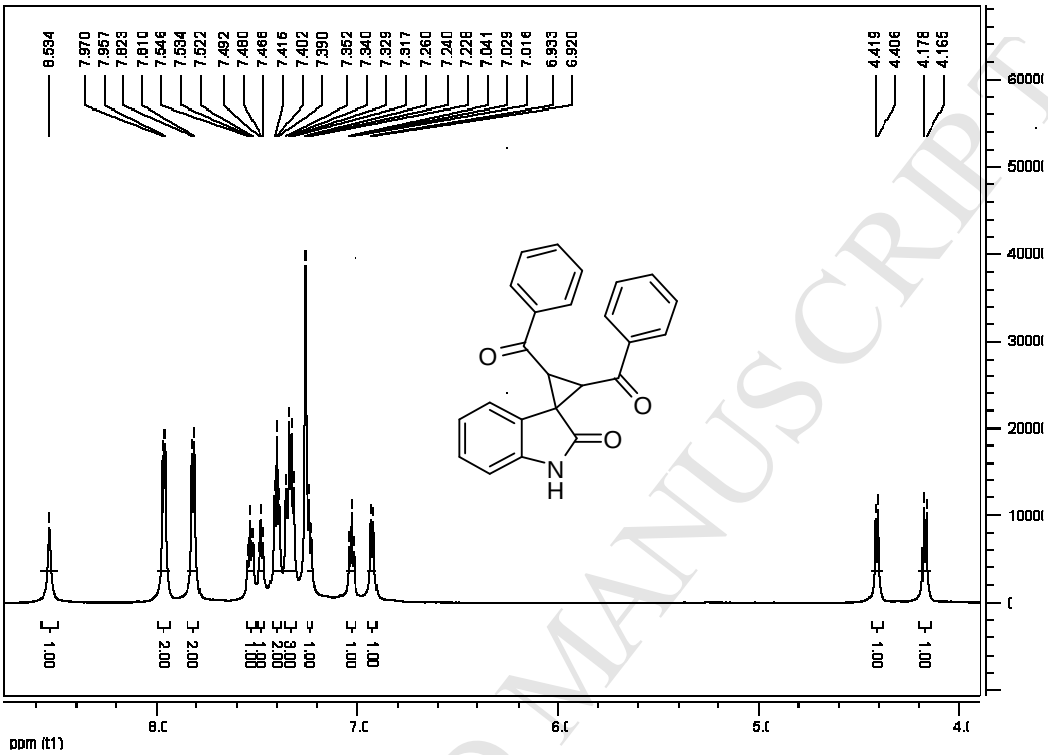


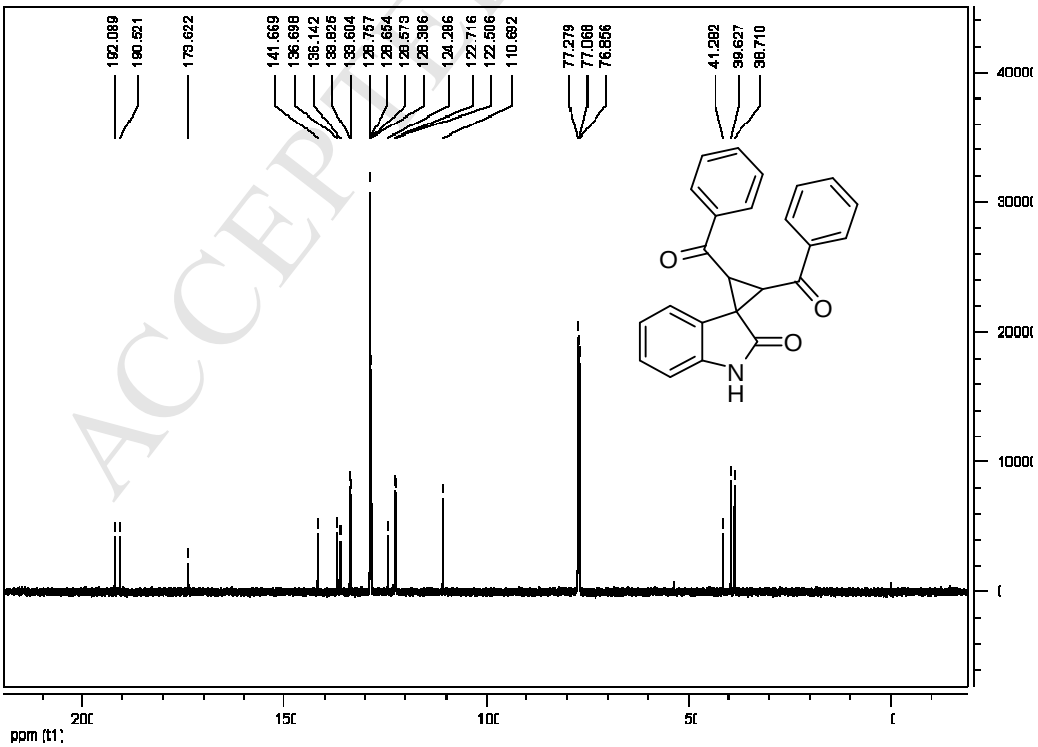
5'-chloro-N,N-diethyl-3-(4-methylbenzoyl)-2'-oxospiro[cyclopropane-1,3'-indoline]-2-carbox amide (3j):



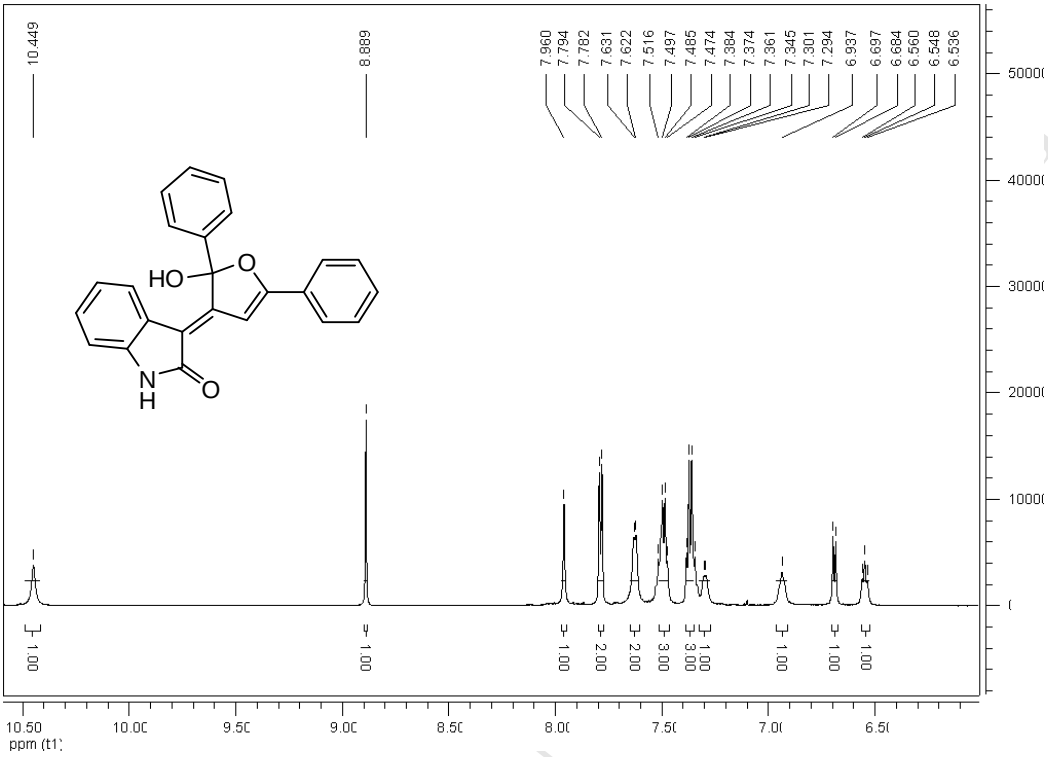


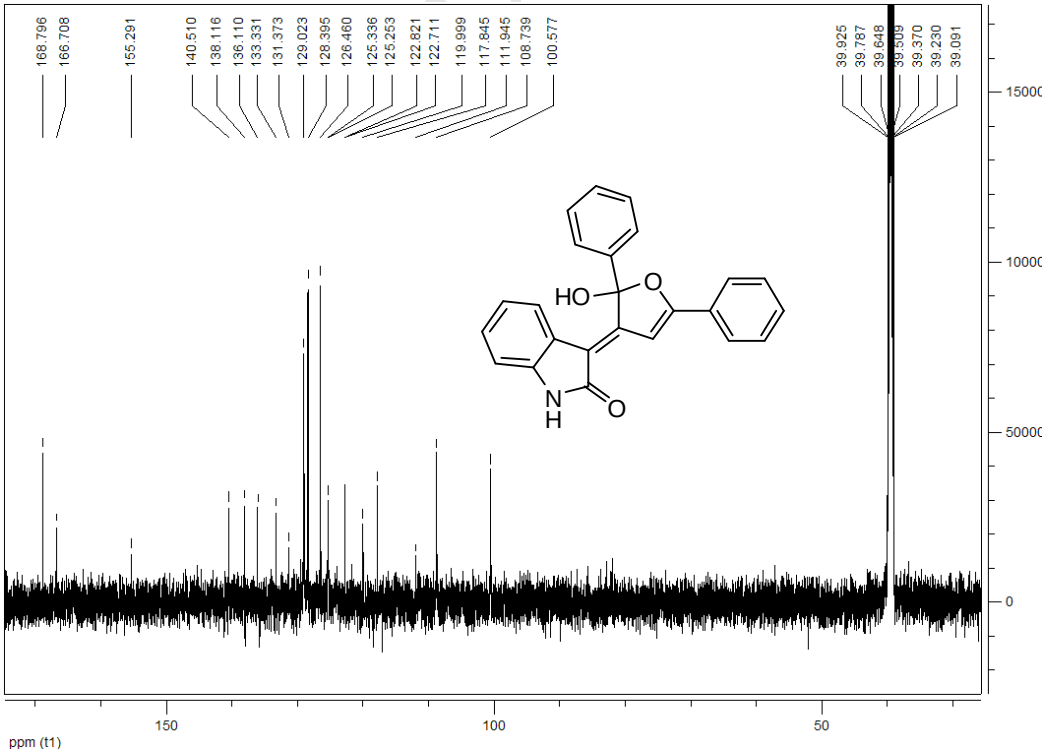
(2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis(phenylmethanone) (4a)：



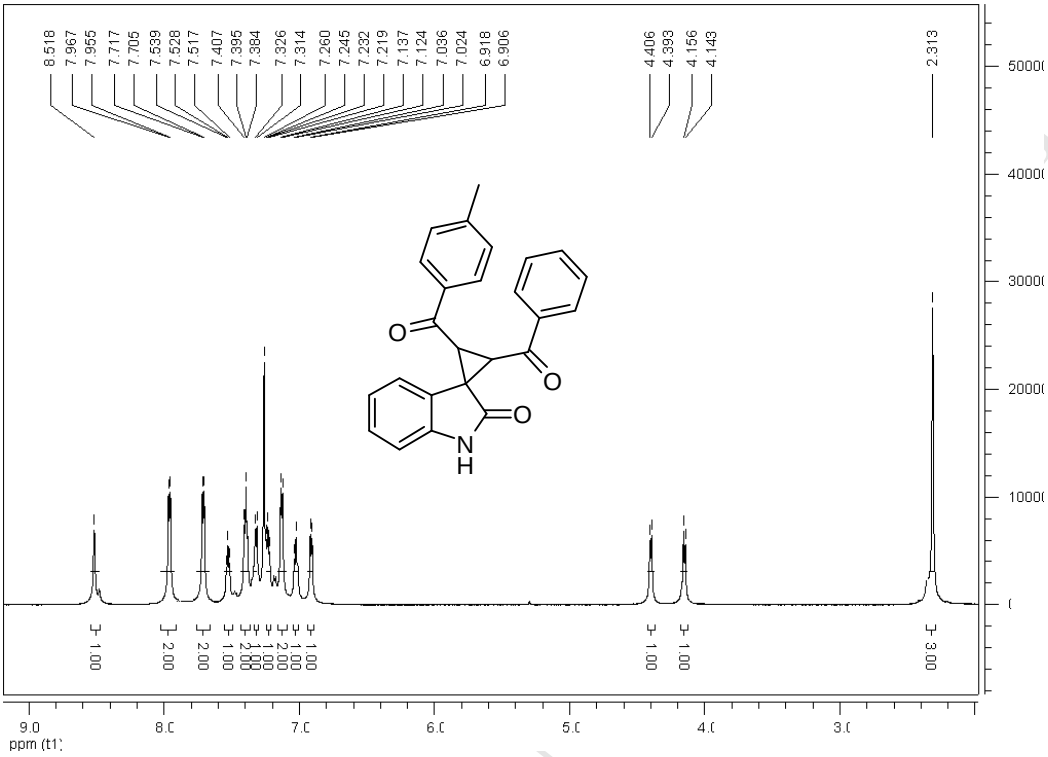


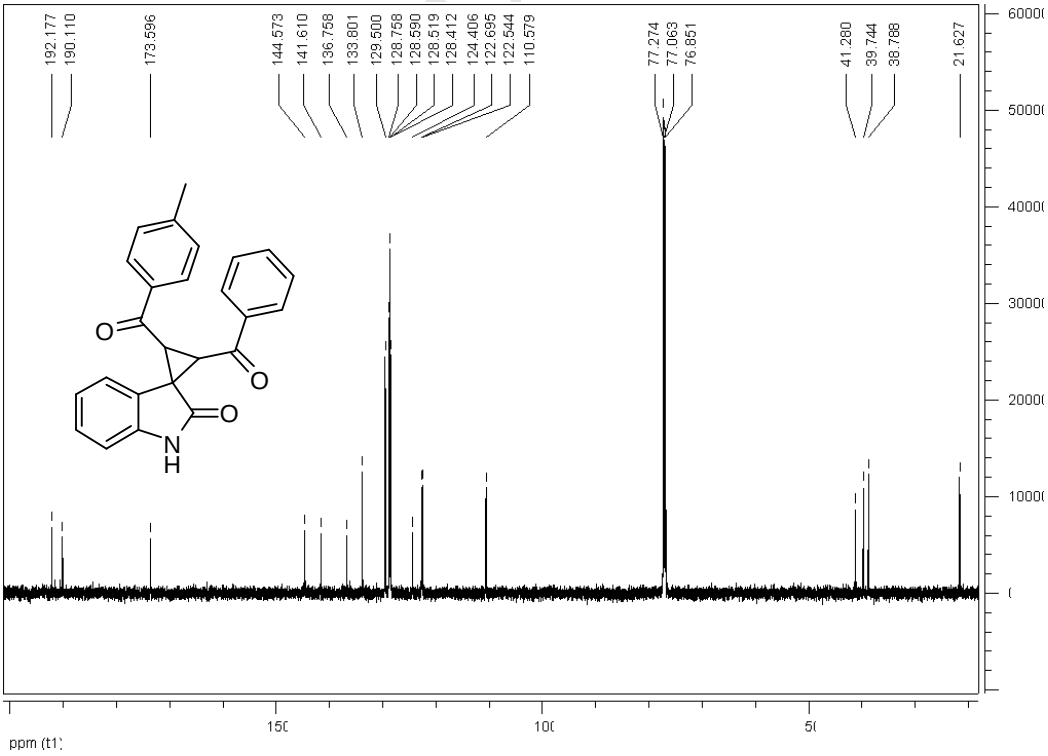
3-(2-hydroxy-2,5-diphenylfuran-3(2H)-ylidene)indolin-2-one 5a:



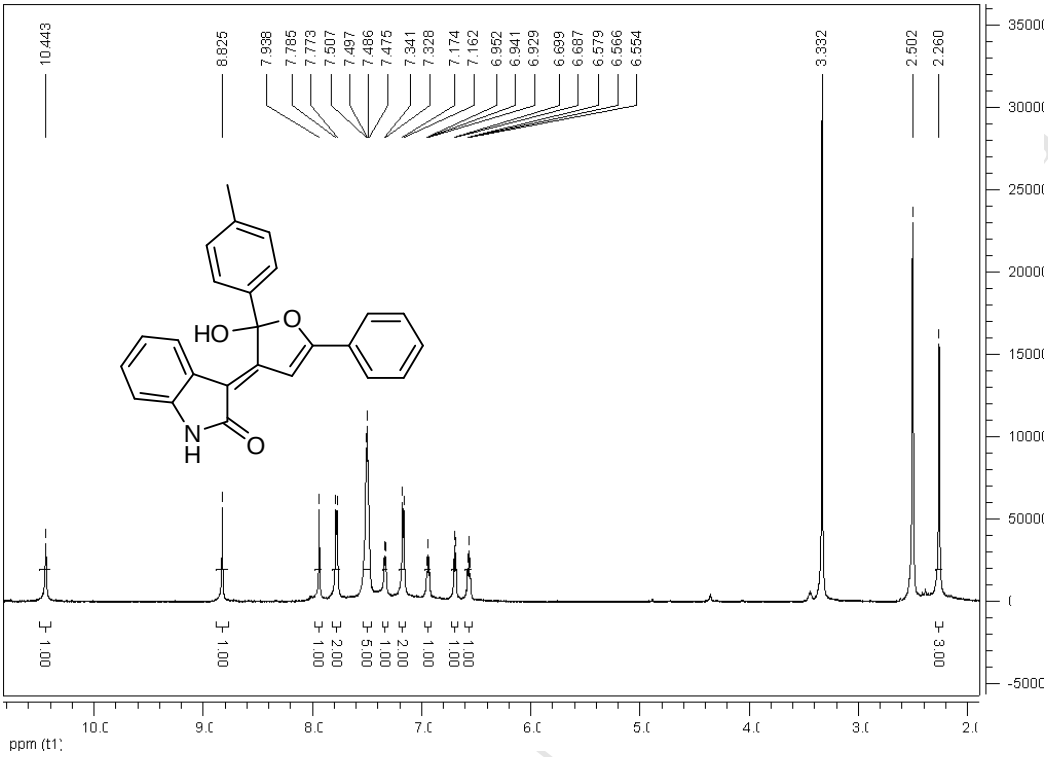


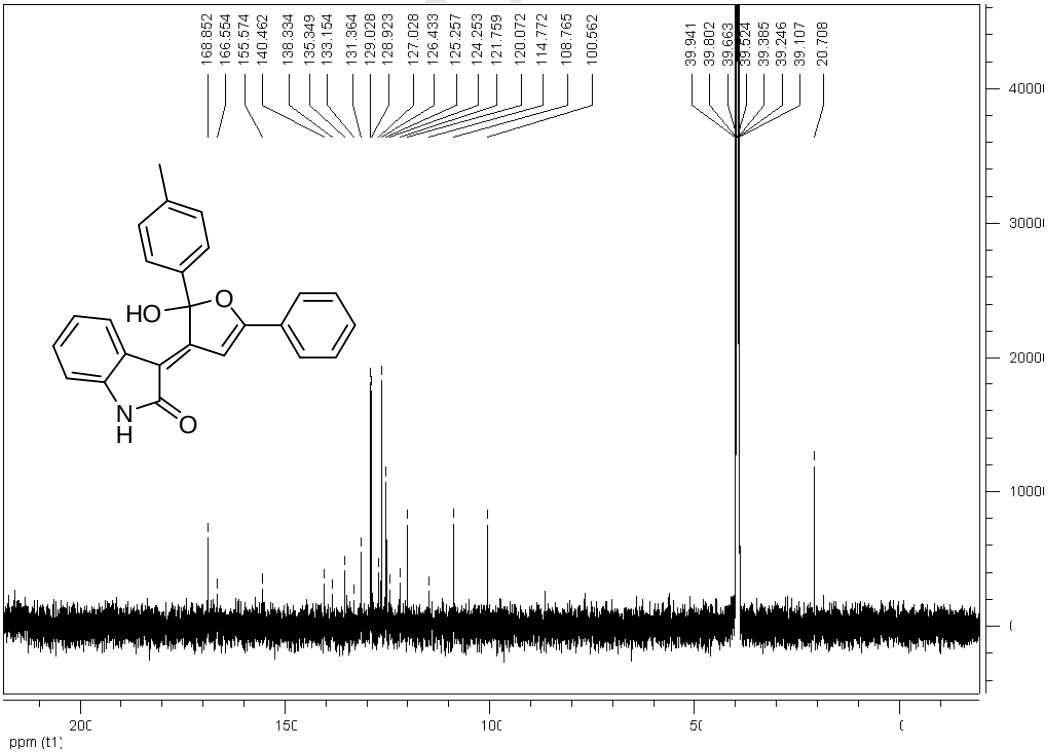
2-benzoyl-3-(4-methylbenzoyl)spiro[cyclopropane-1,3'-indolin]-2'-one (4b):



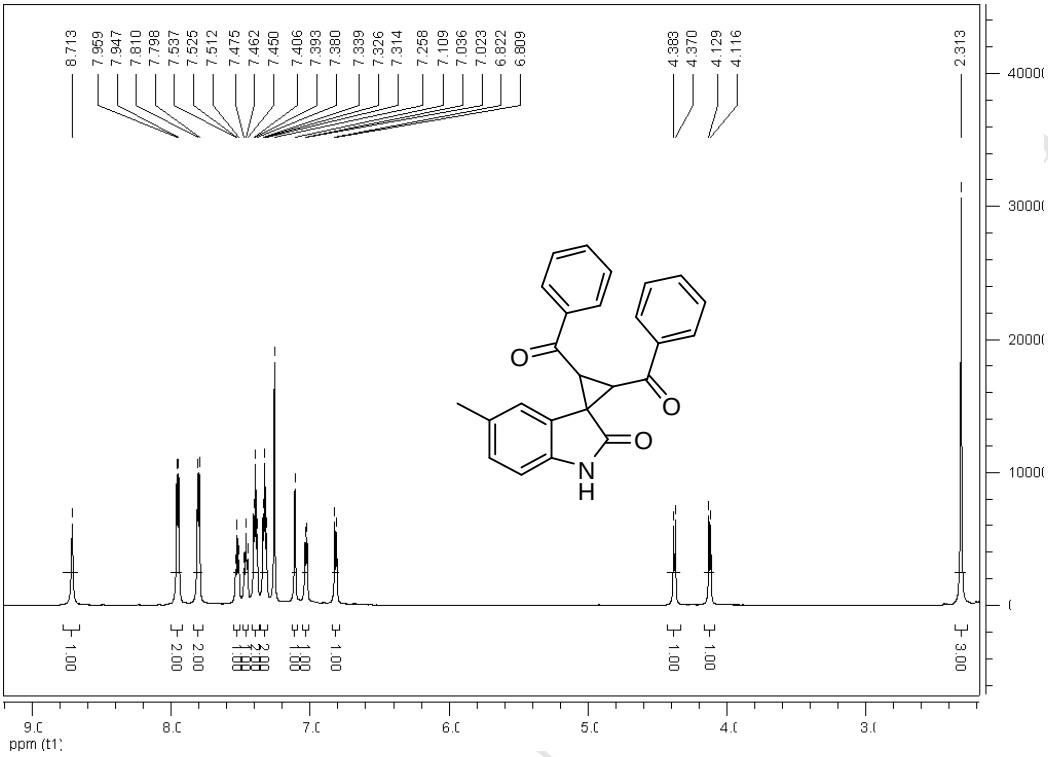


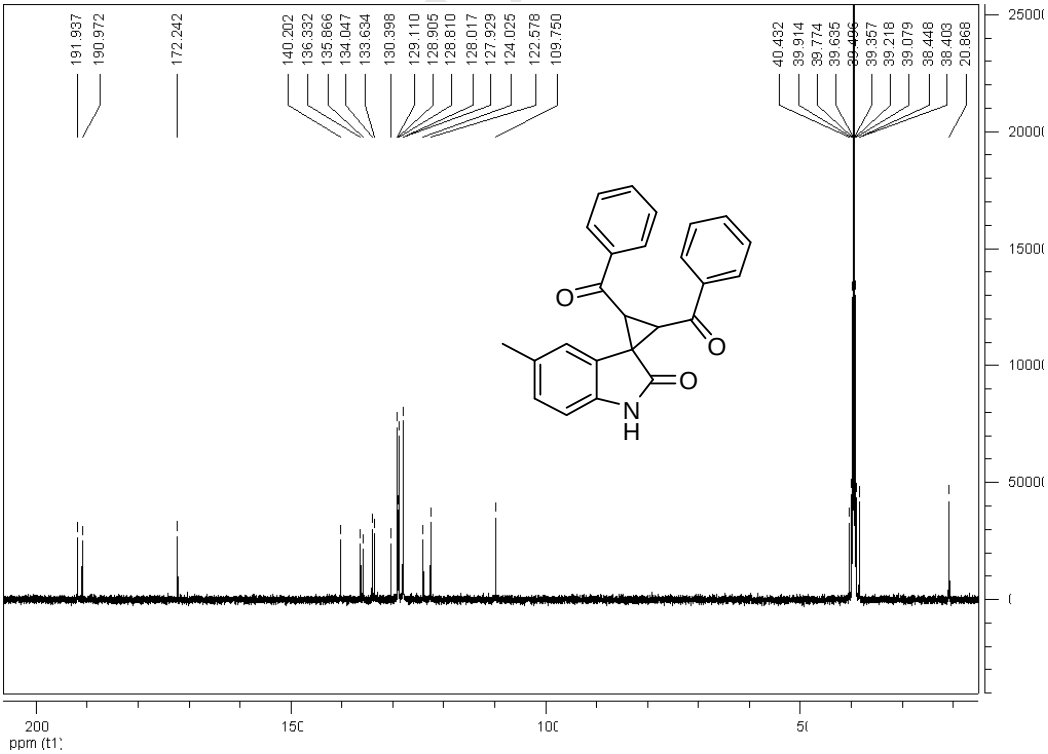
3-(2-hydroxy-5-phenyl-2-p-tolylfuran-3(2H)-ylidene)indolin-2-one (5b):



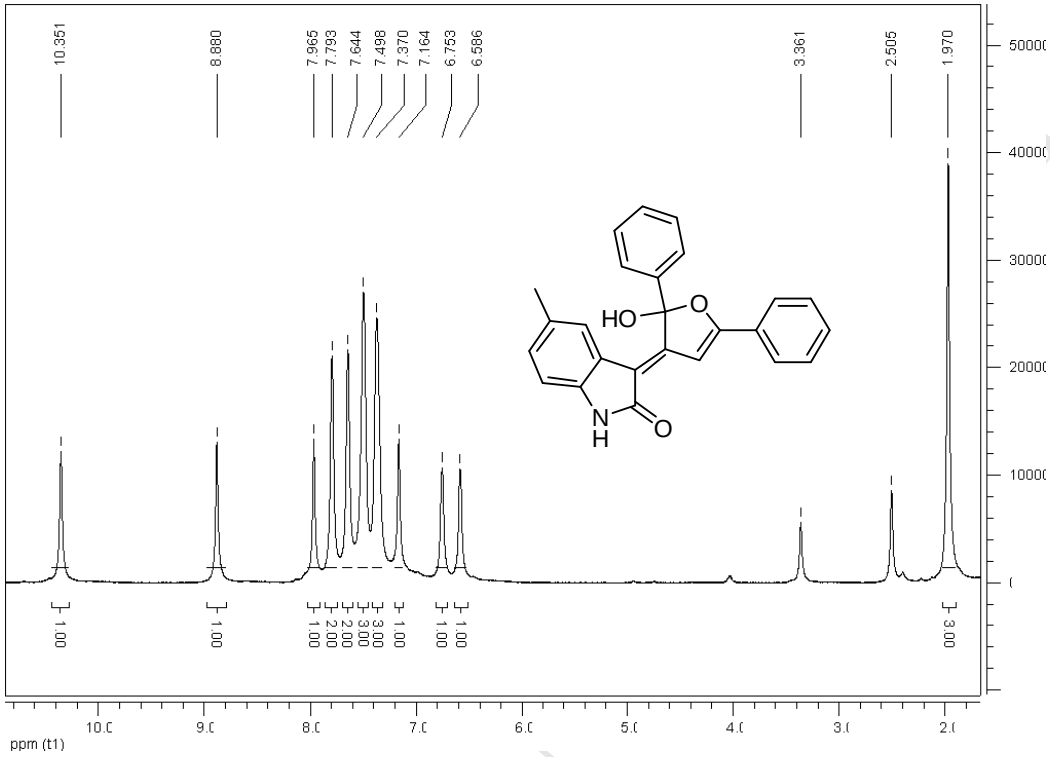


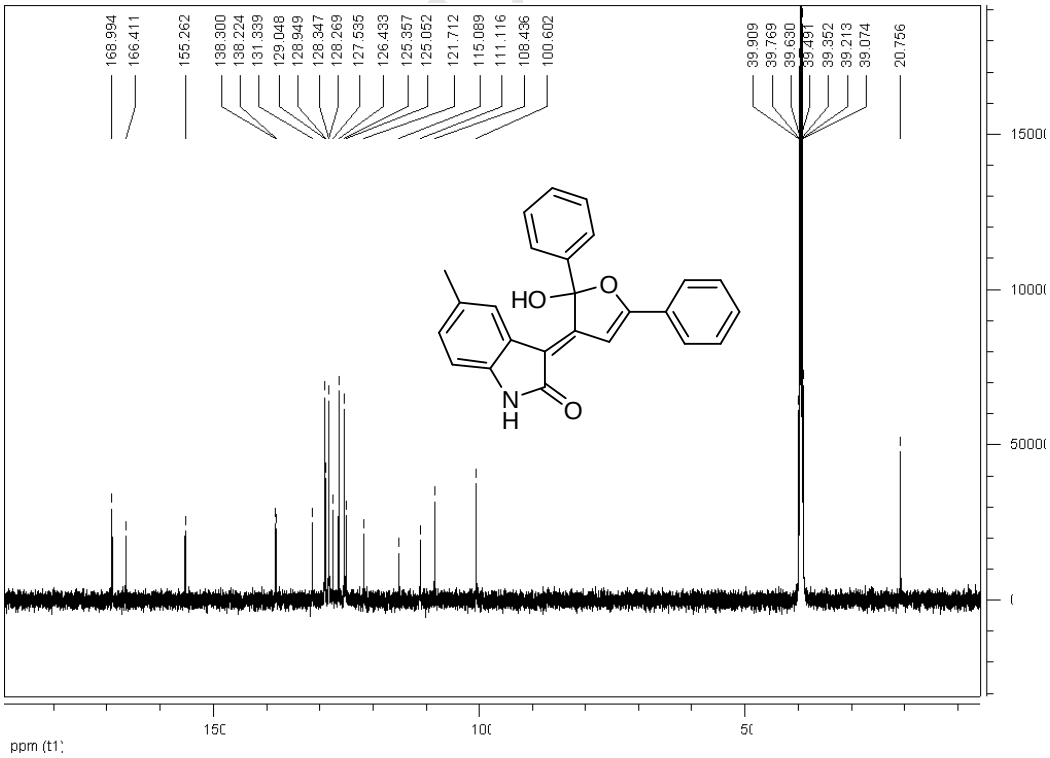
(5'-methyl-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl) bis(phenyl-methanone) (4c):



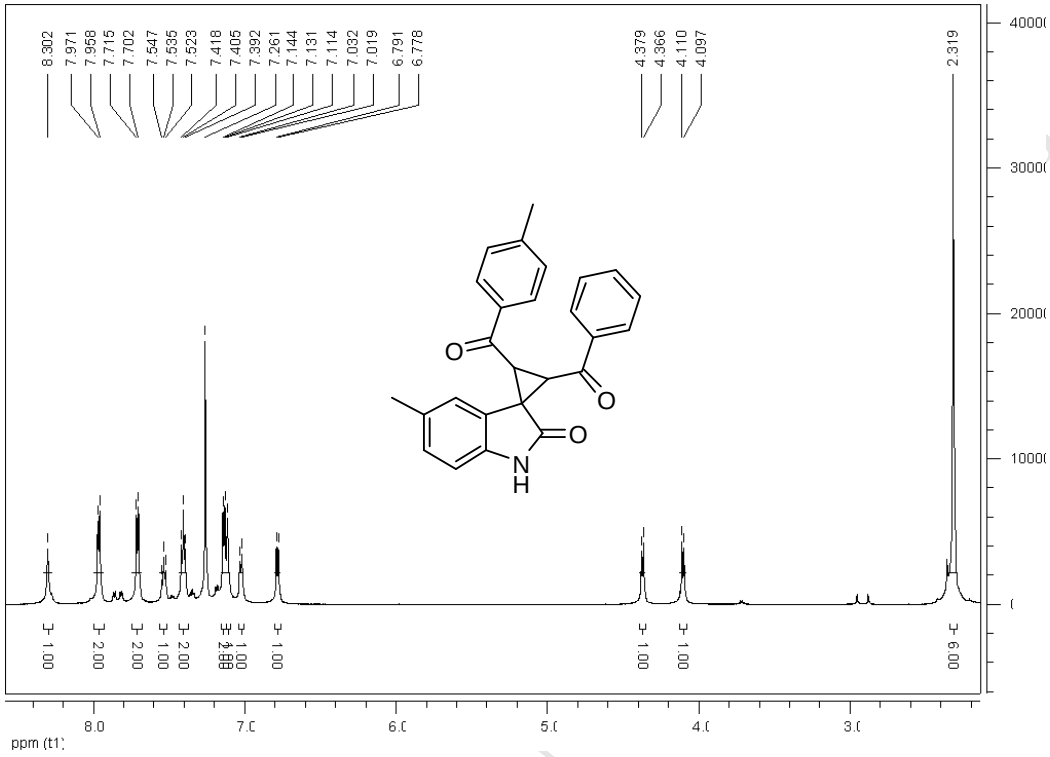


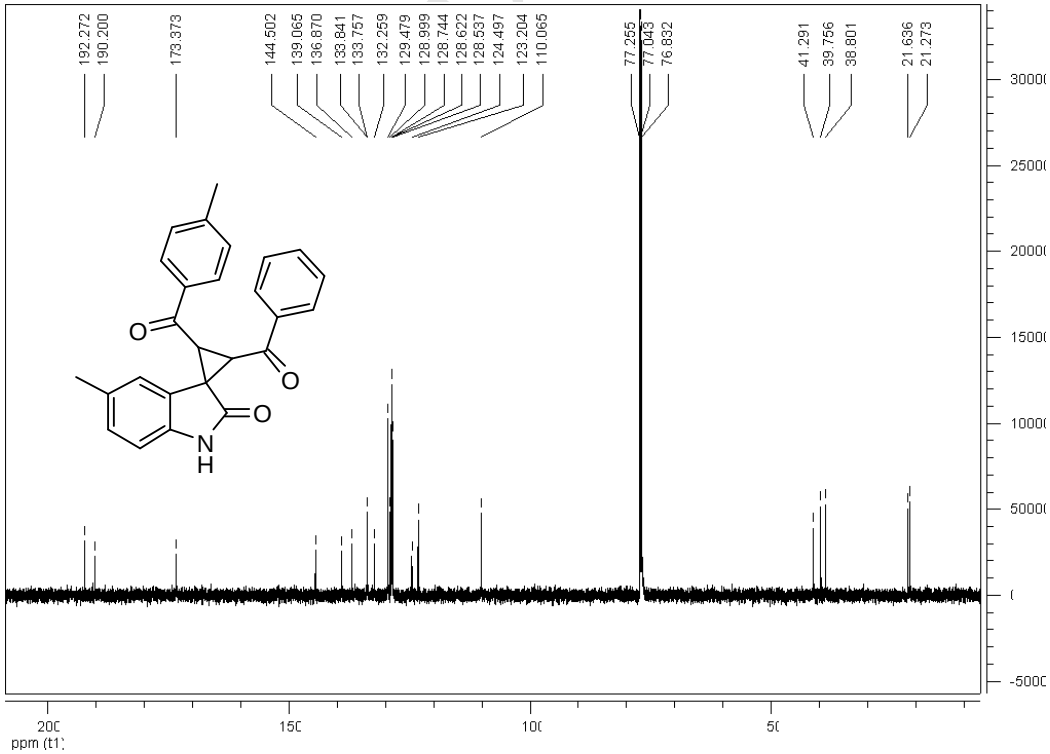
3-(2-hydroxy-2,5-diphenylfuran-3(2H)-ylidene)-5-methylindolin-2-one (5c):



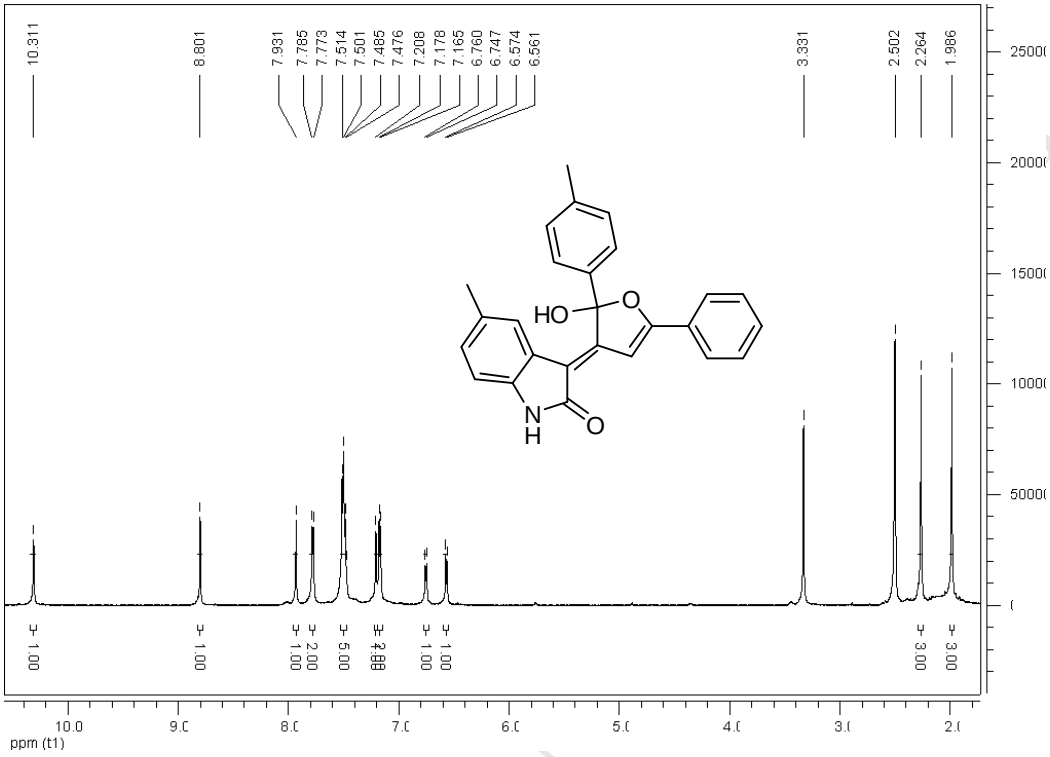


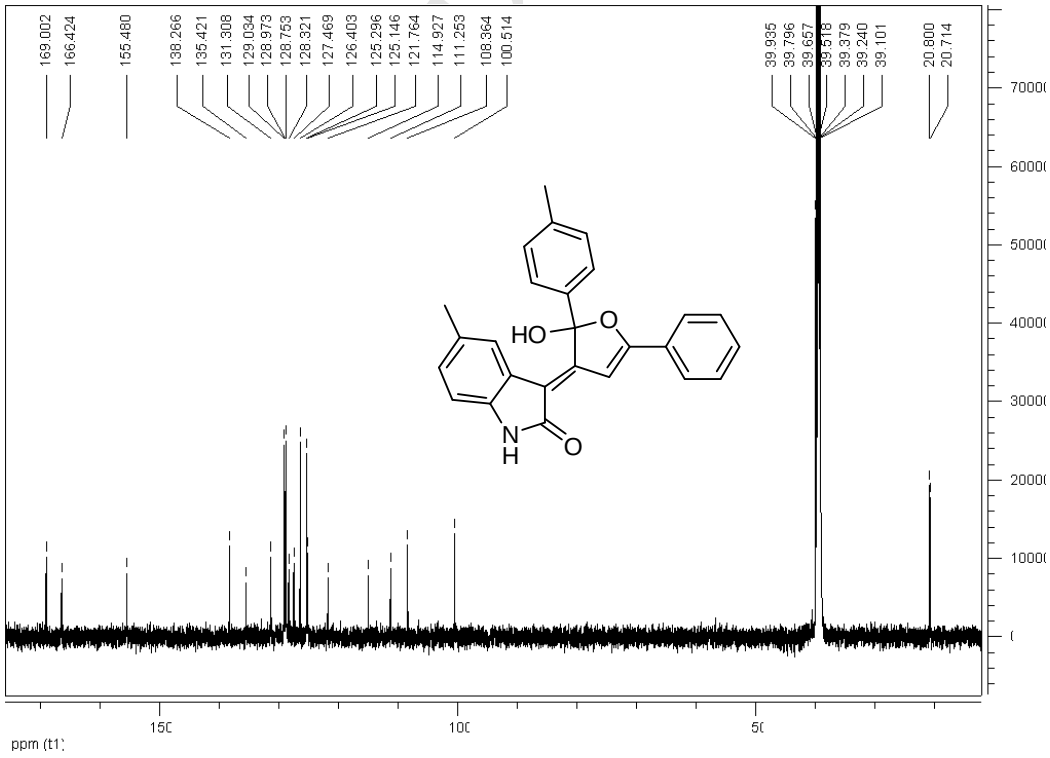
2-benzoyl-5'-methyl-3-(4-methylbenzoyl)spiro[cyclopropane-1,3'-indolin]-2'- one (4d):



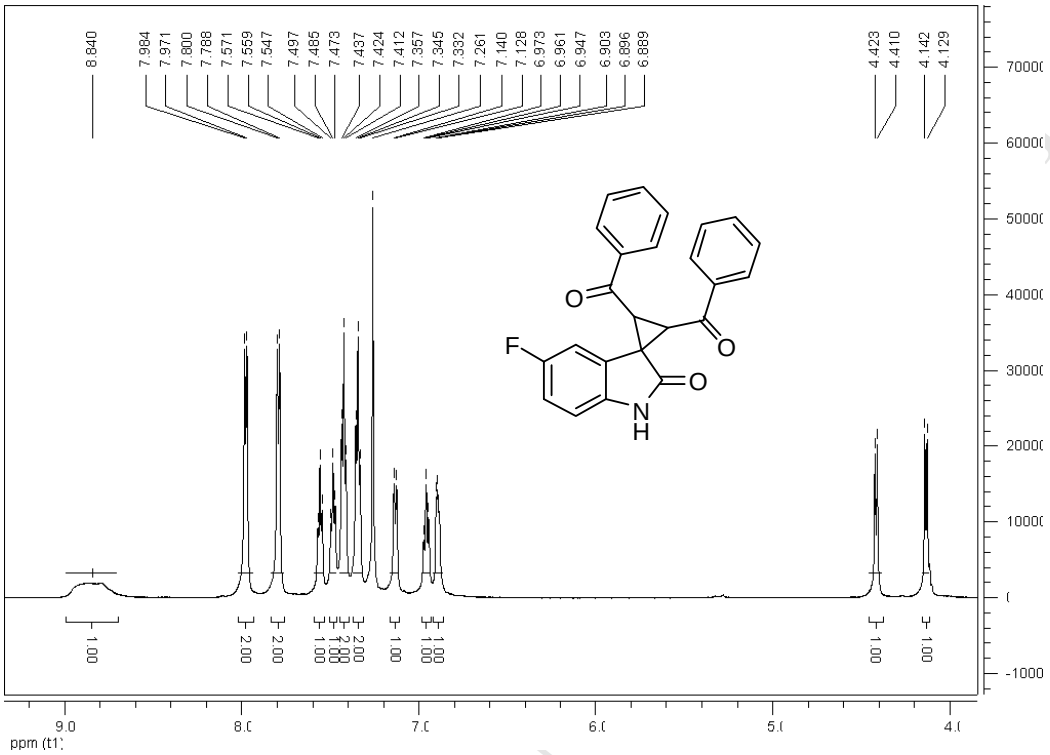


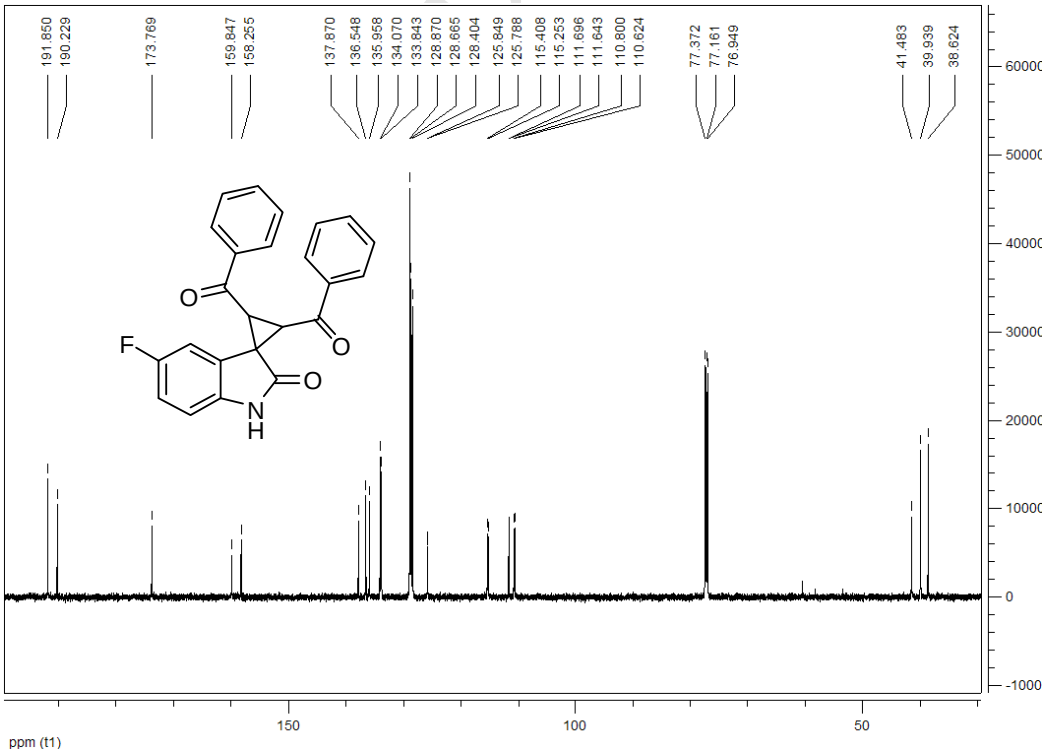
3-(2-hydroxy-5-phenyl-2-p-tolylfuran-3(2H)-ylidene)-5-methylindolin-2-one (5d):



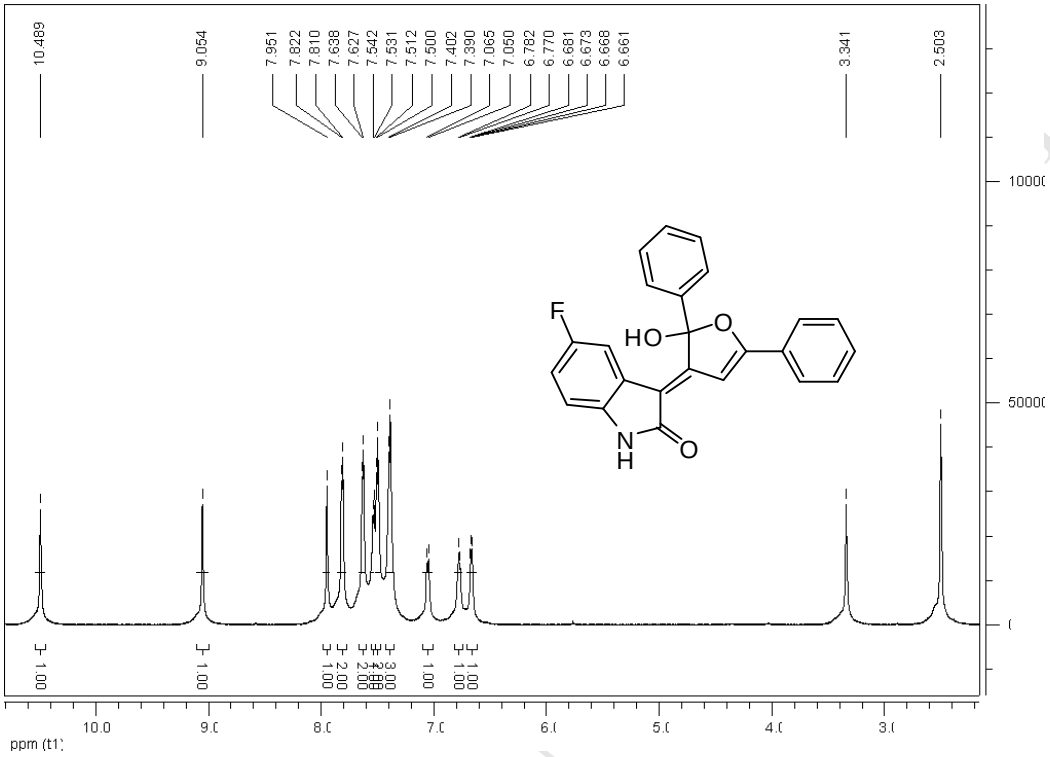


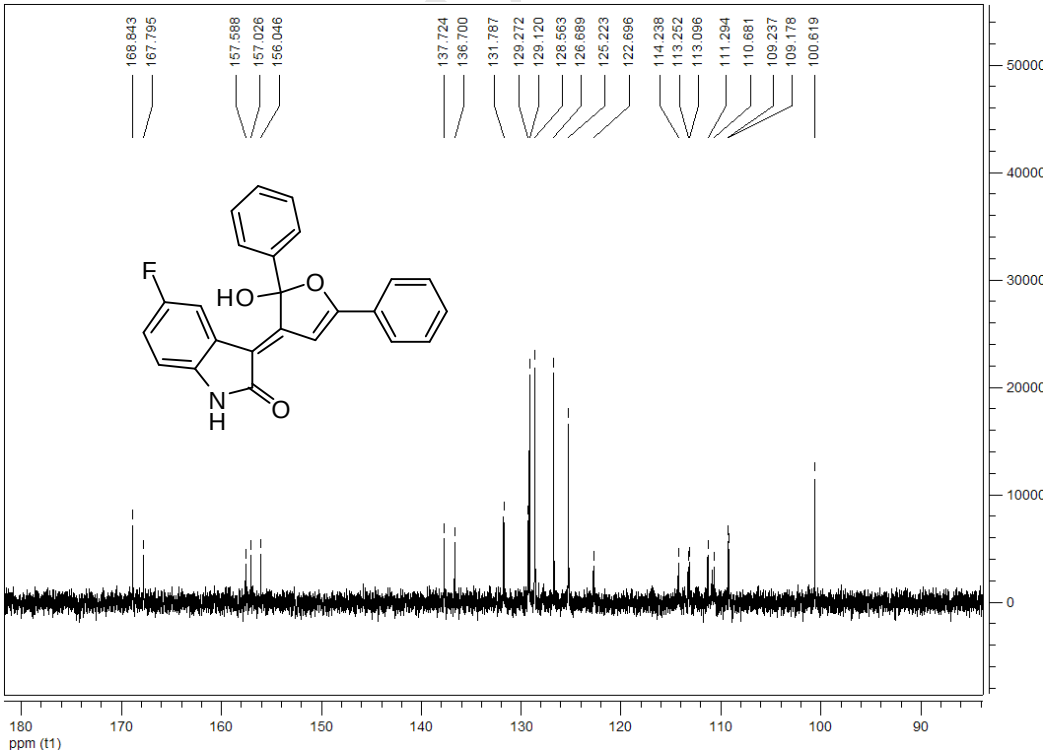
(5'-fluoro-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis(phenylmethan-one) (4e):



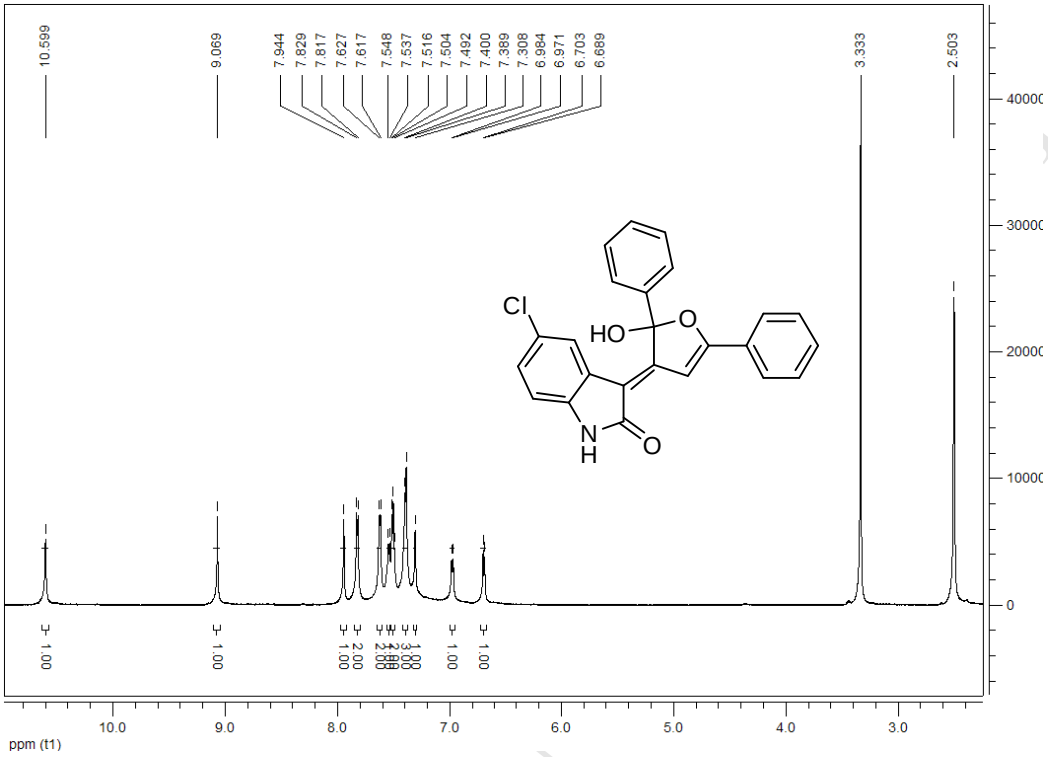


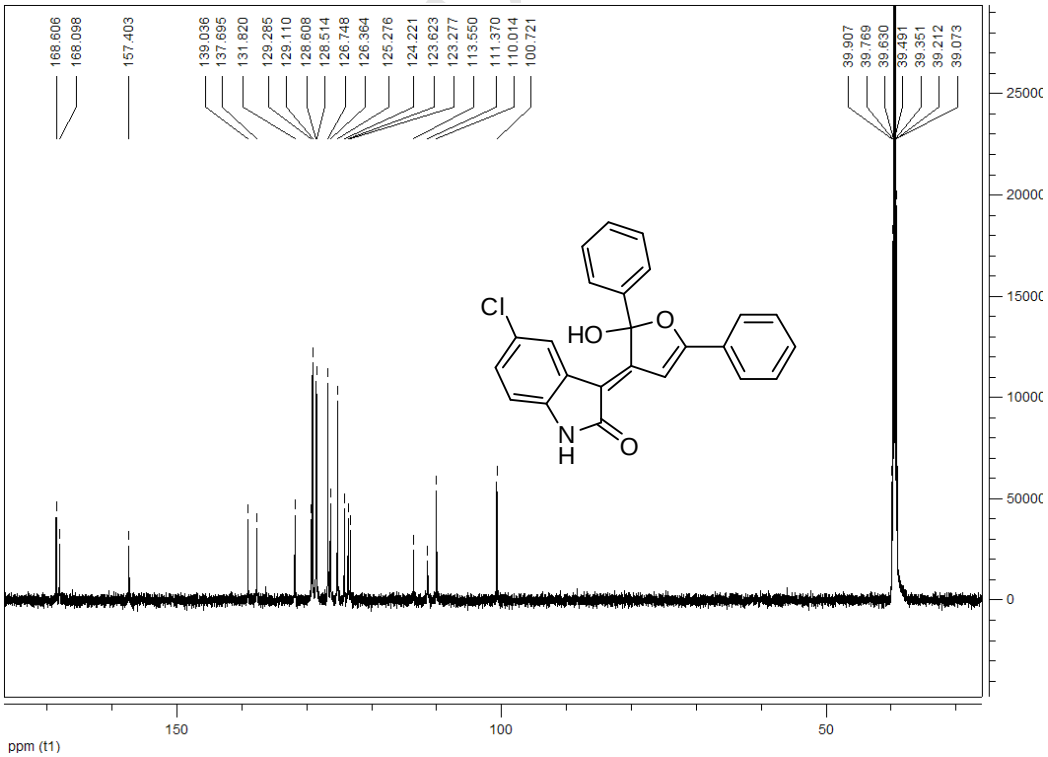
5-fluoro-3-(2-hydroxy-2,5-diphenylfuran-3(2H)-ylidene)indolin-2-one (5e):



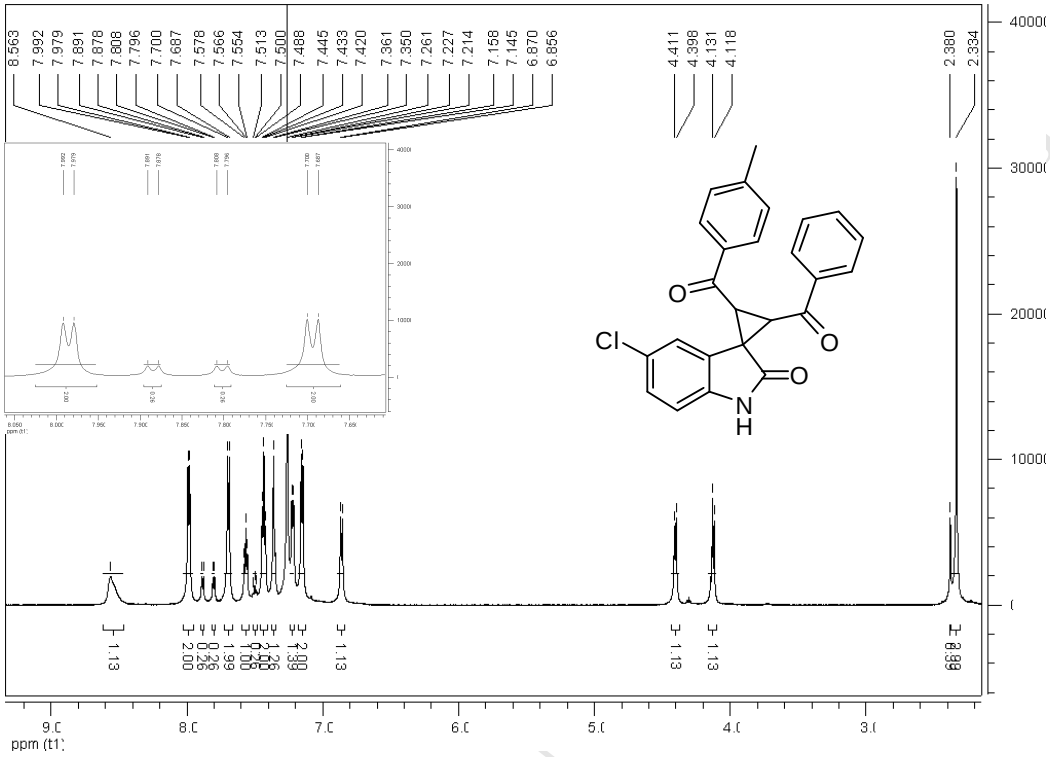


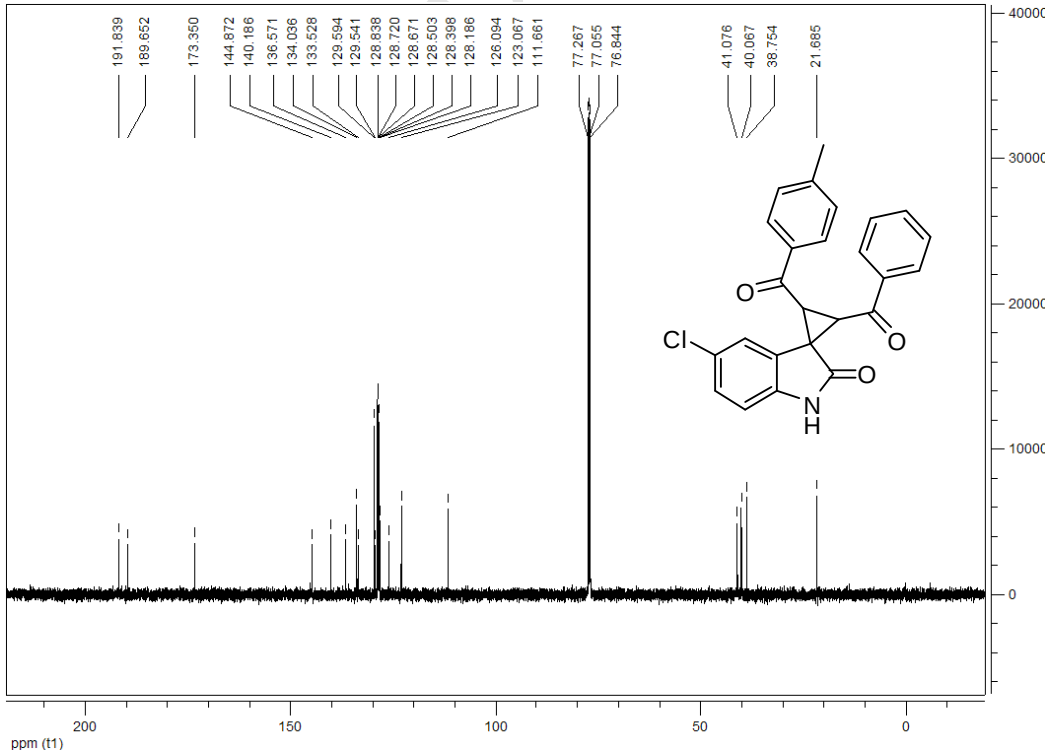
5-chloro-3-(2-hydroxy-2,5-diphenylfuran-3(2H)-ylidene)indolin-2-one (5f):



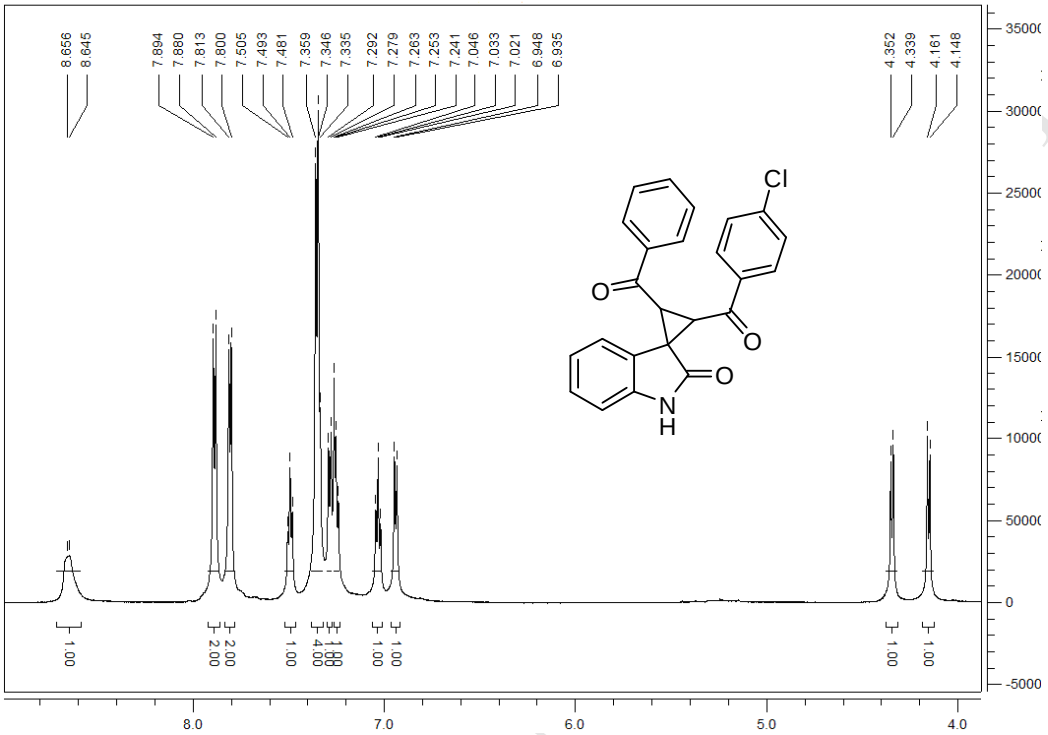


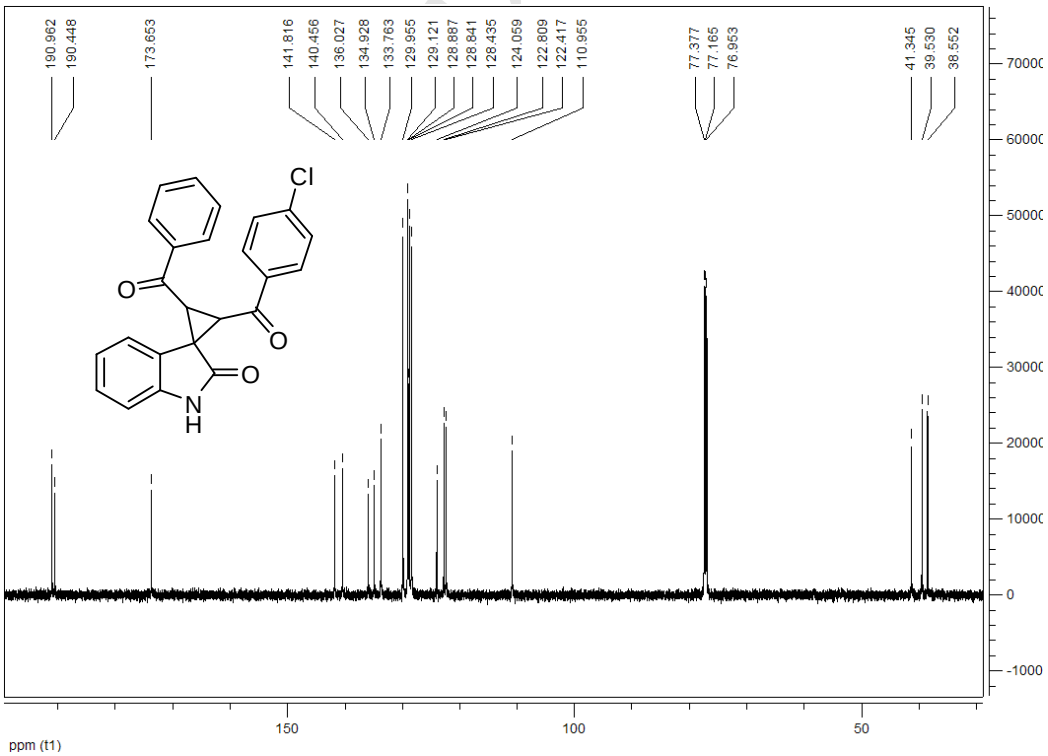
2-benzoyl-5'-chloro-3-(4-methylbenzoyl)spiro[cyclopropane-1,3'-indolin]- 2'-one (4g):



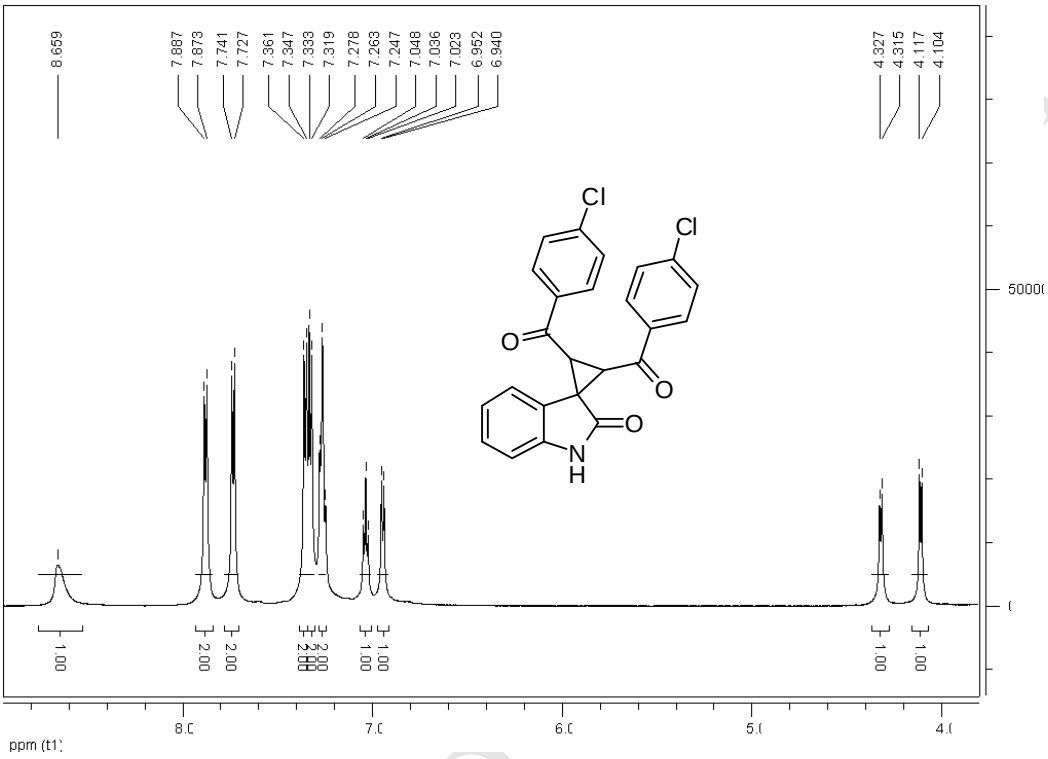


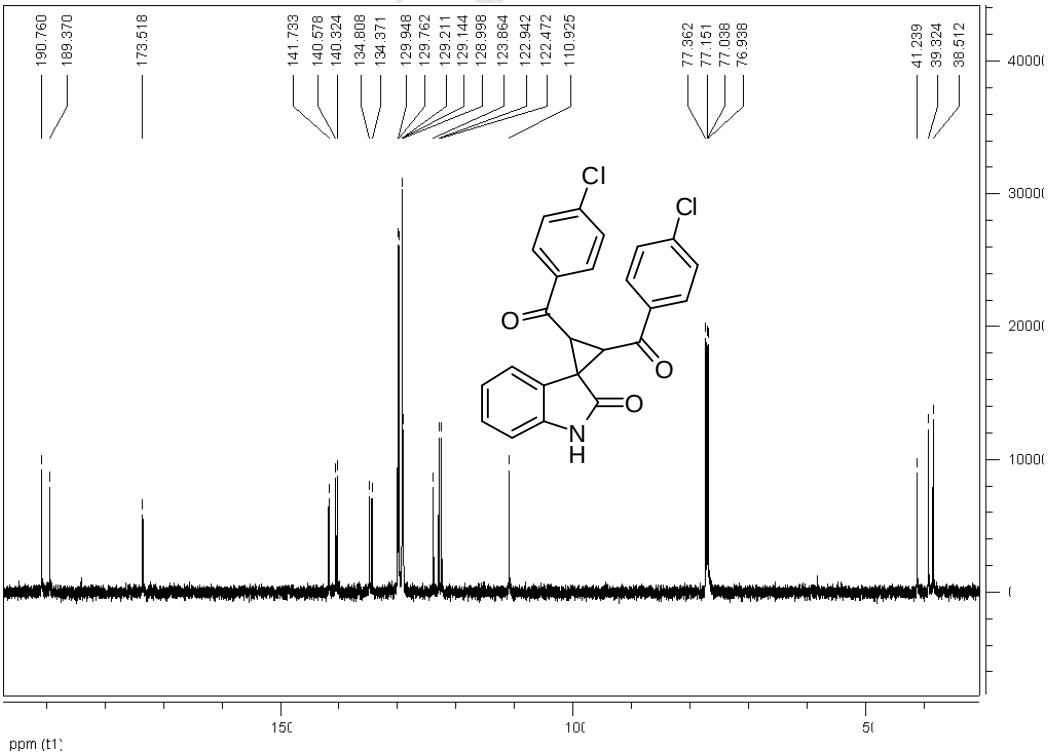
2-benzoyl-3-(4-chlorobenzoyl)spiro[cyclopropane-1,3'-indolin]-2'-one (4h):



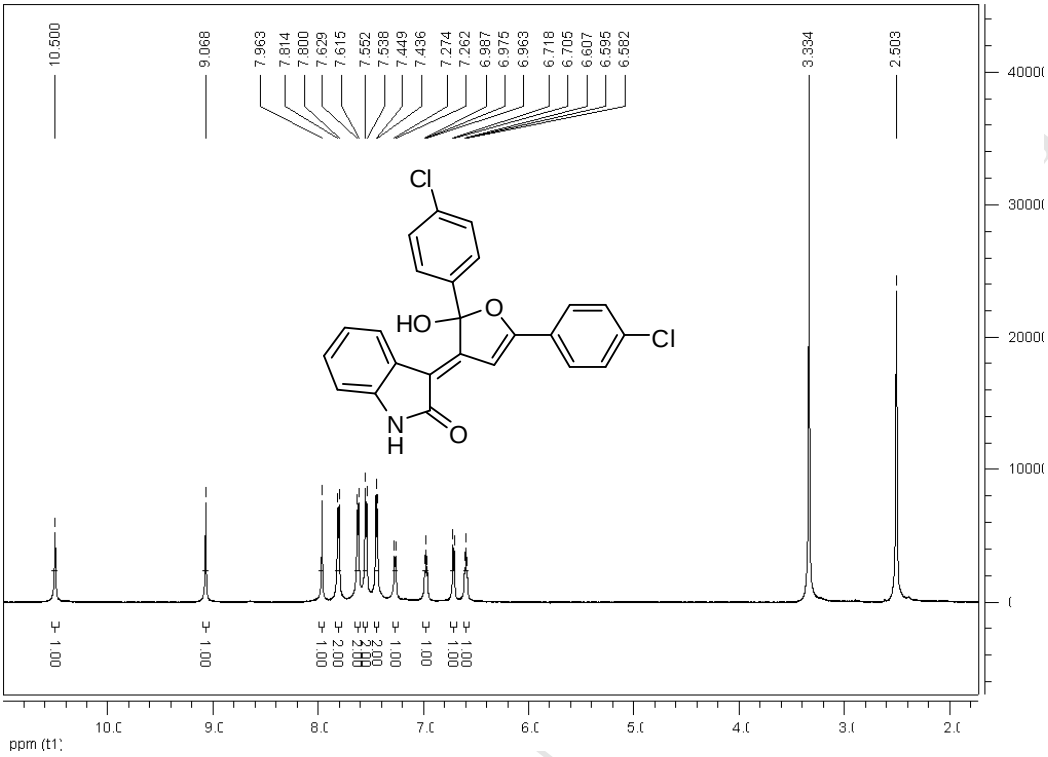


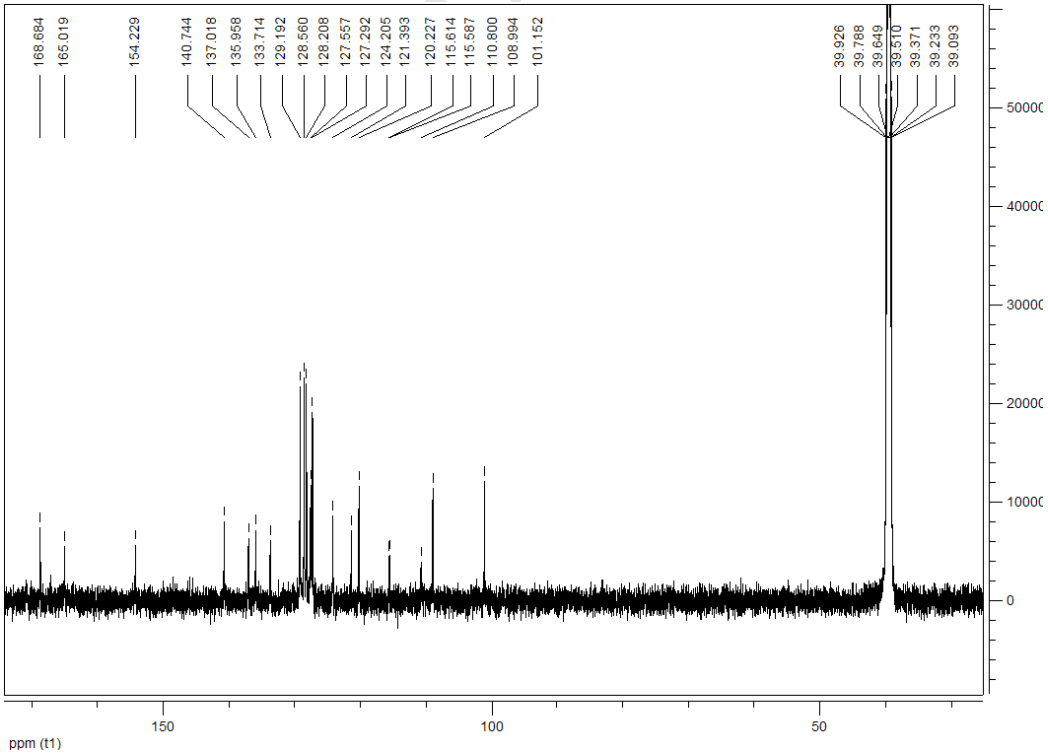
(2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl) bis((4-chloroph enyl) methanone) (4i)：



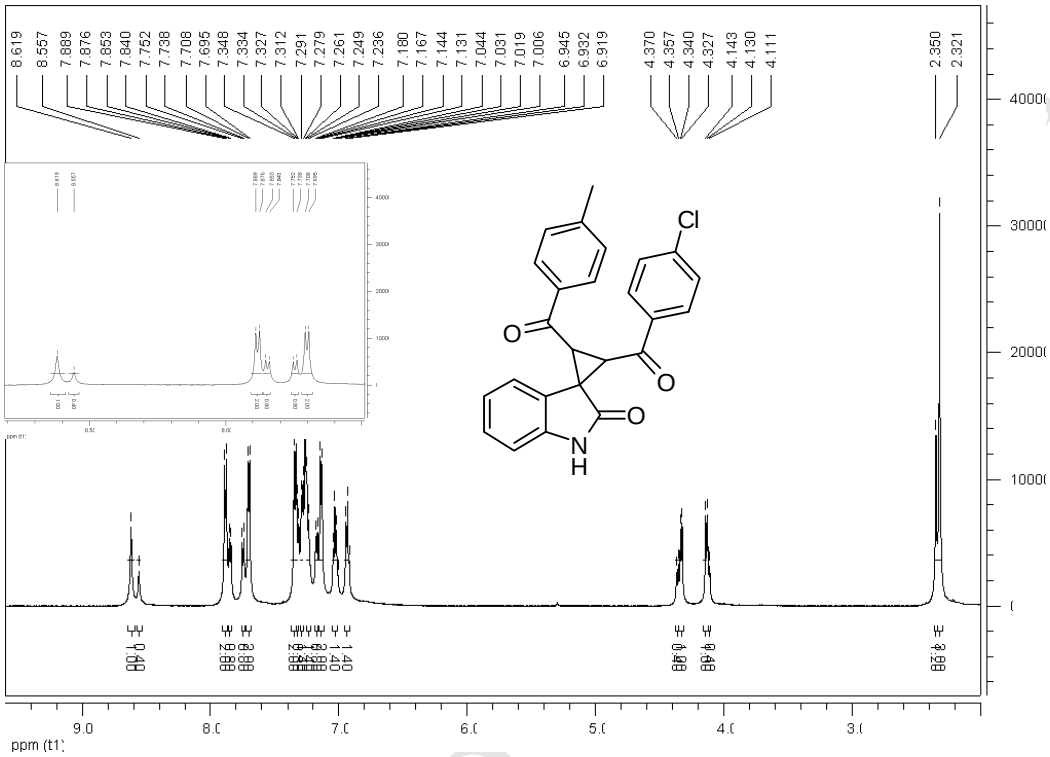


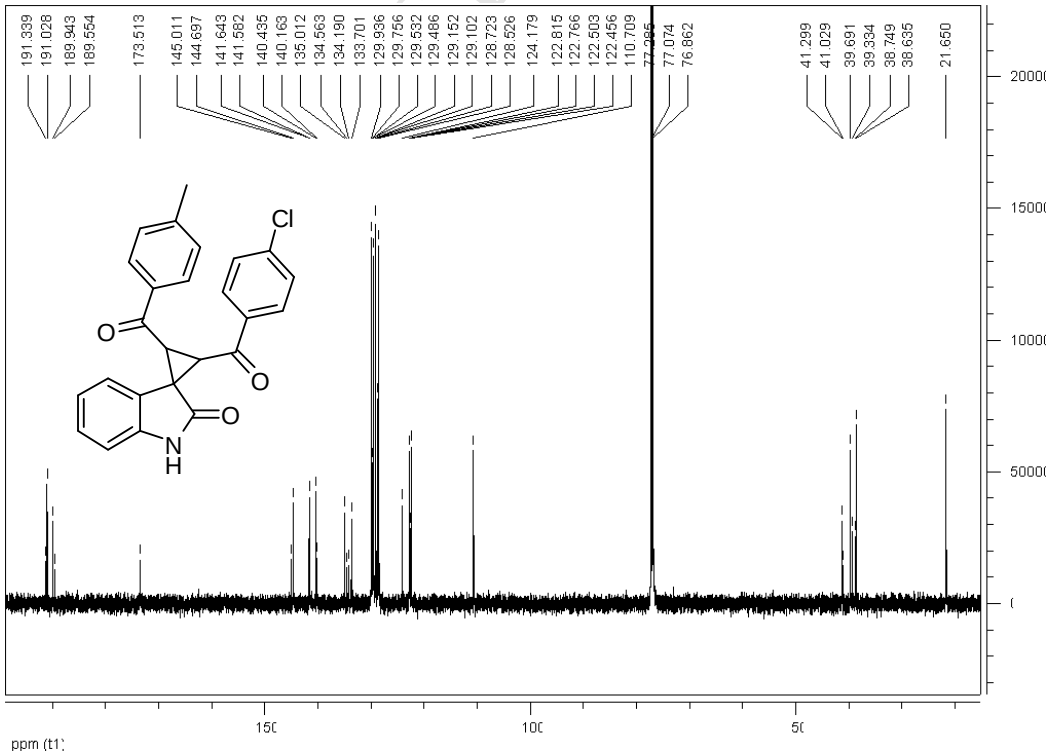
3-(2,5-bis(4-chlorophenyl)-2-hydroxyfuran-3(2H)-ylidene)indolin-2-one (5i):



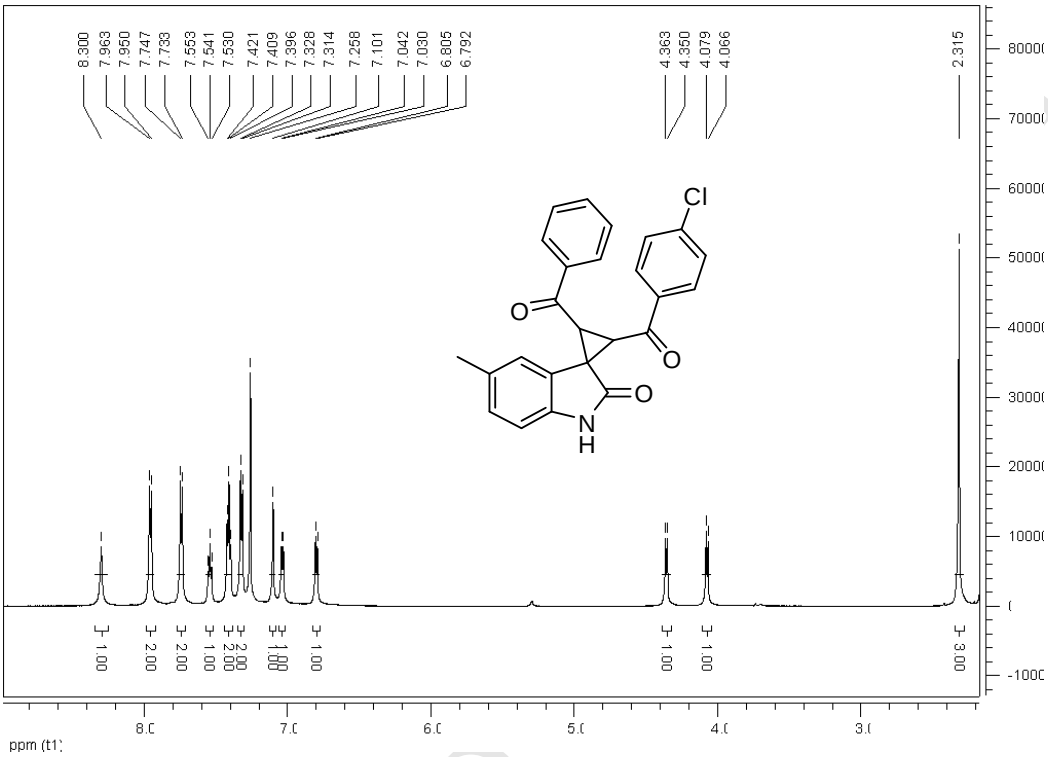


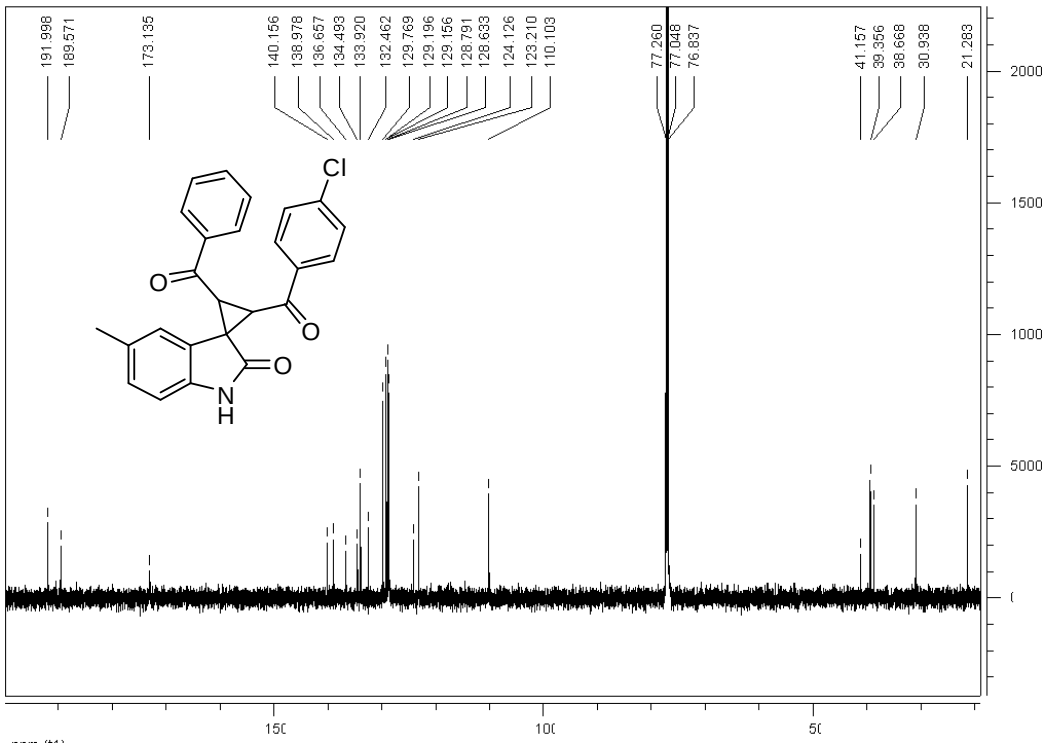
2-(4-chlorobenzoyl)-3-(4-methylbenzoyl)2-(4-chlorobenzoyl)-3-(4-methyl-benzoyl)spiro[cyclo -propane-1,3'-indolin]-2'-one (4j):



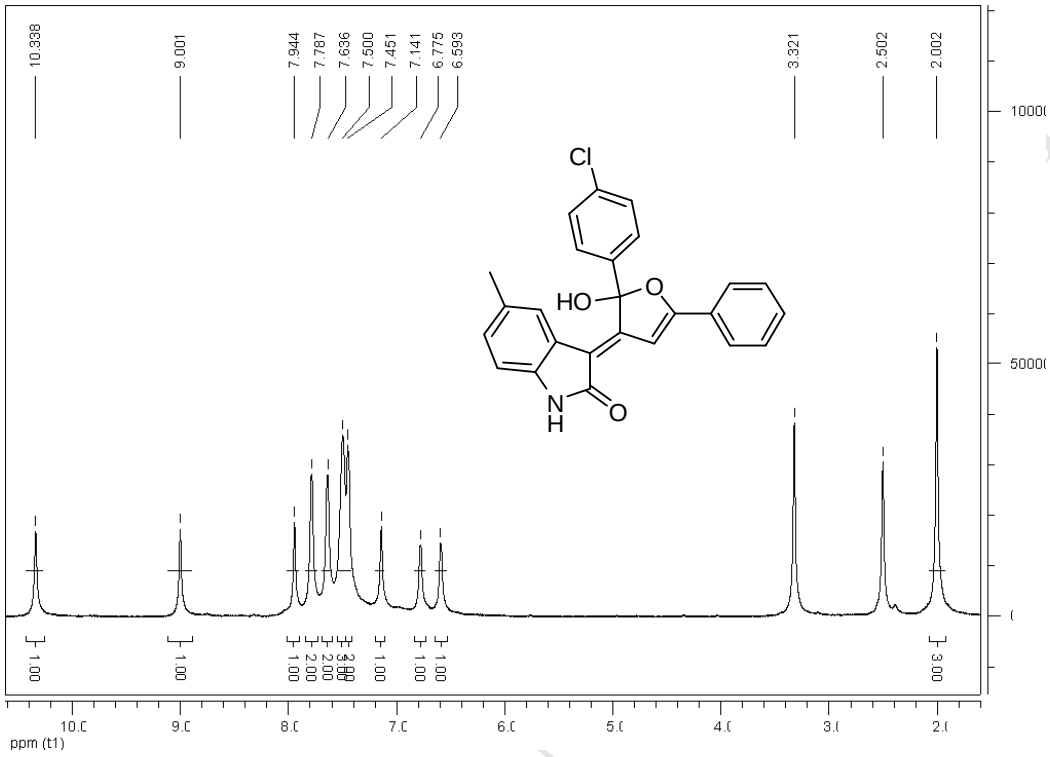


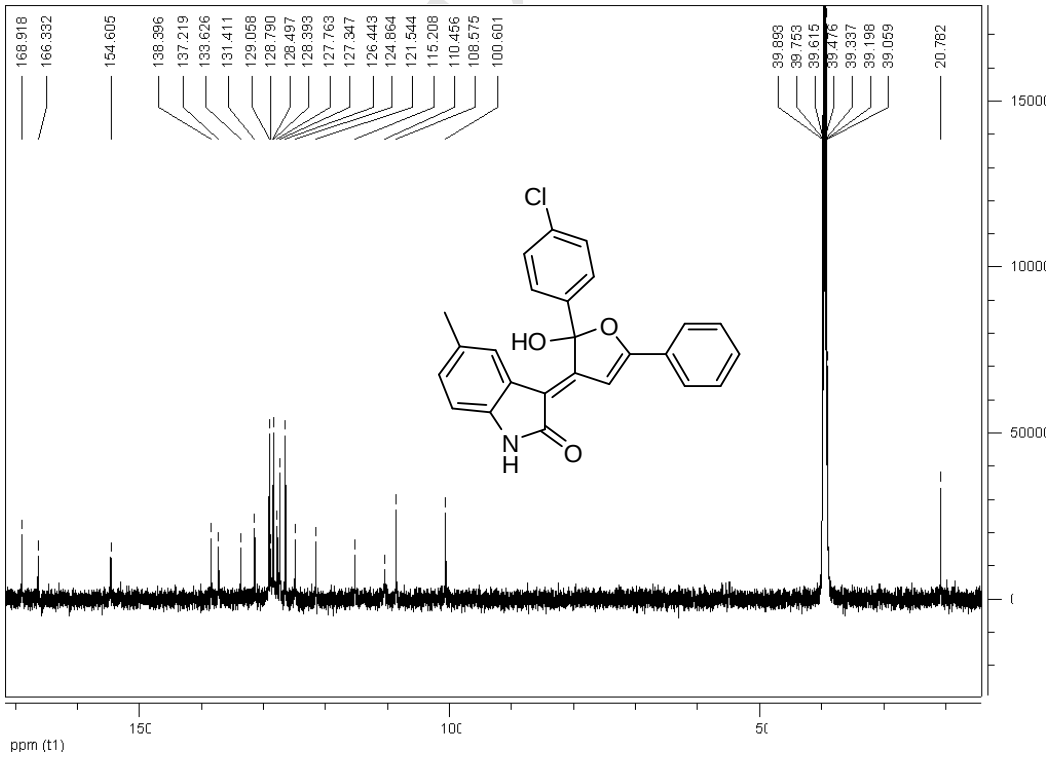
2-benzoyl-3-(4-chlorobenzoyl)-5'-methylspiro[cyclopropane-2-benzoyl-3-(4-chlorobenzoyl)-5'-methylspiro[cyclopropane-1,3'-indolin]-2'-one (4k):



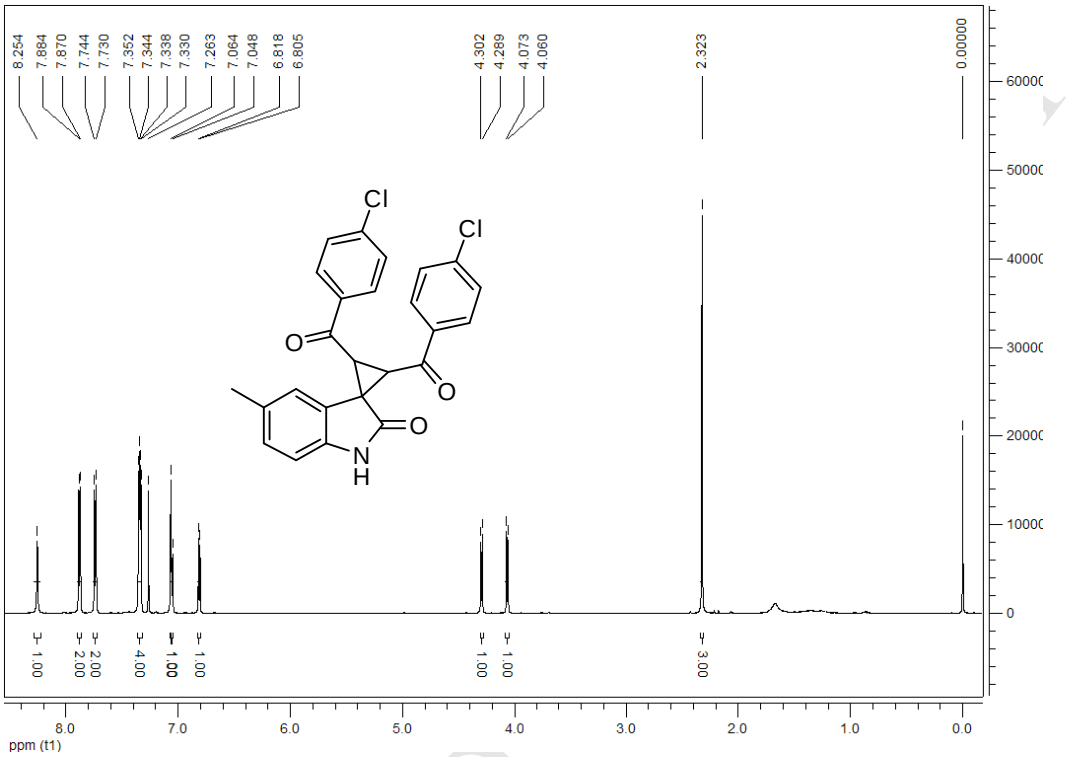


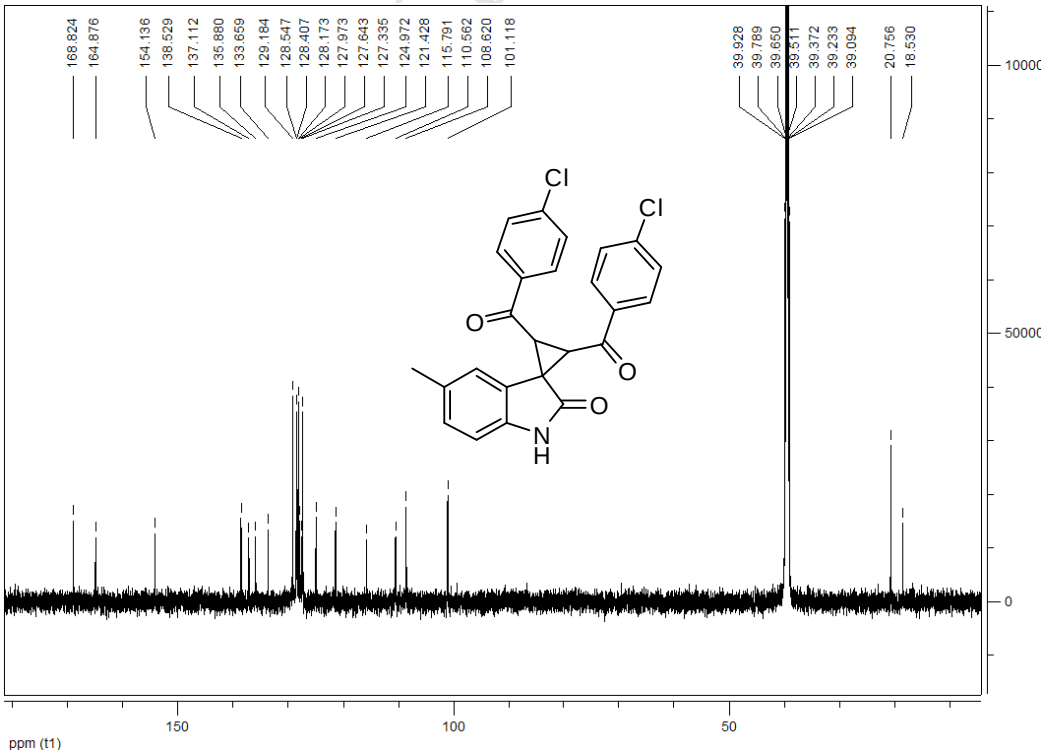
3-(2-(4-chlorophenyl)-2-hydroxy-5-phenylfuran-3(2H)-ylidene)-5-methylindol-in-2-one (5k):



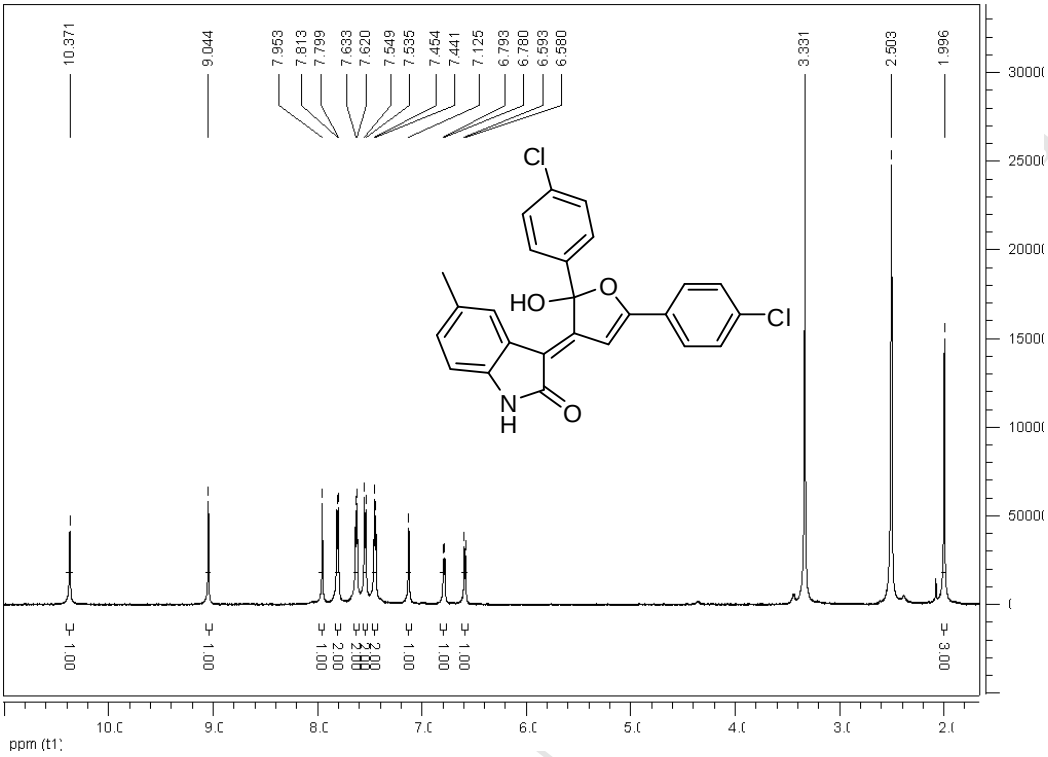


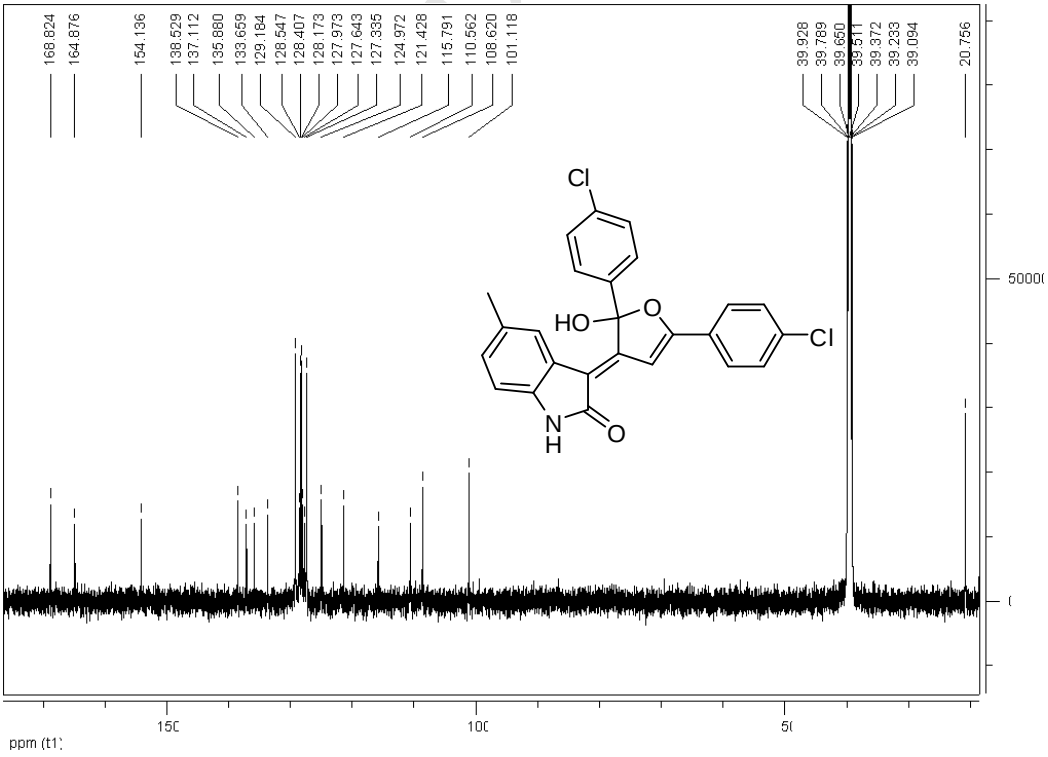
(5'-methyl-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis((4-chloro- phenyl)methanone) (4l):



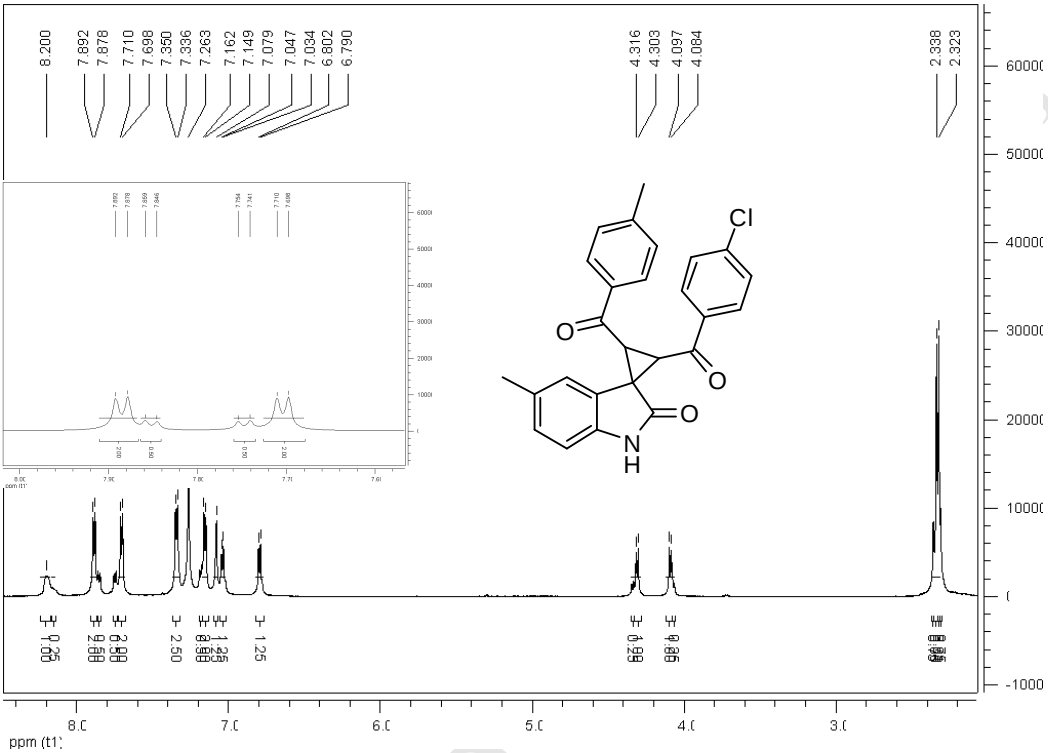


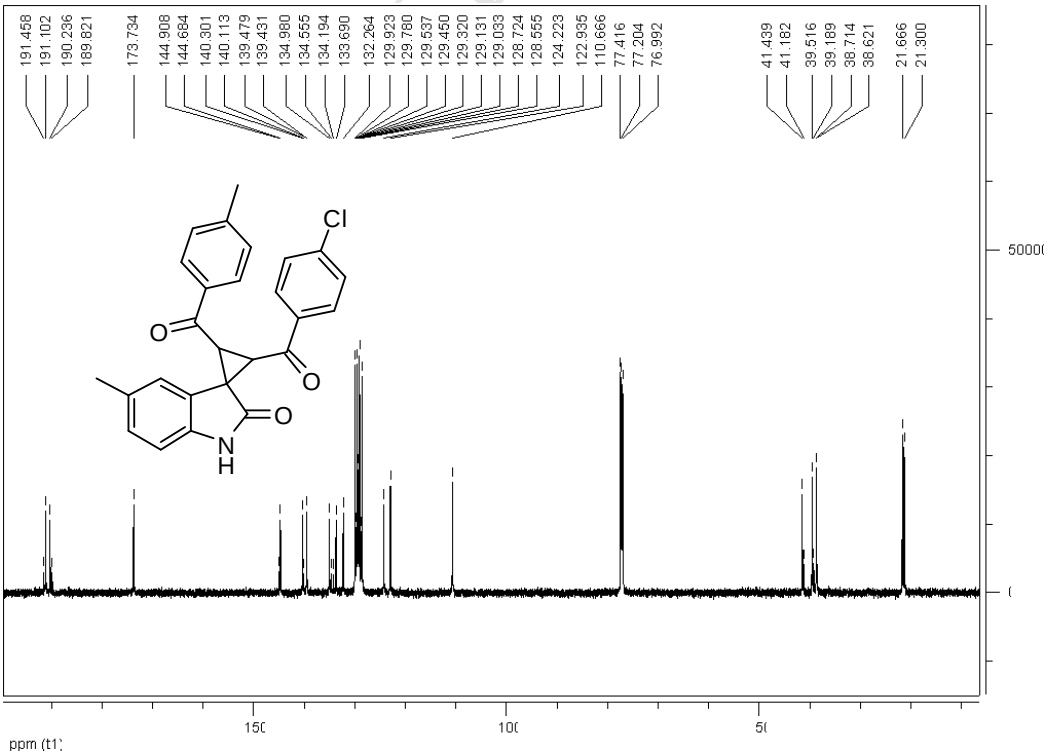
3-(2,5-bis(4-chlorophenyl)-2-hydroxyfuran-3(2H)-ylidene)-5-methylindolin-2-one (5l):



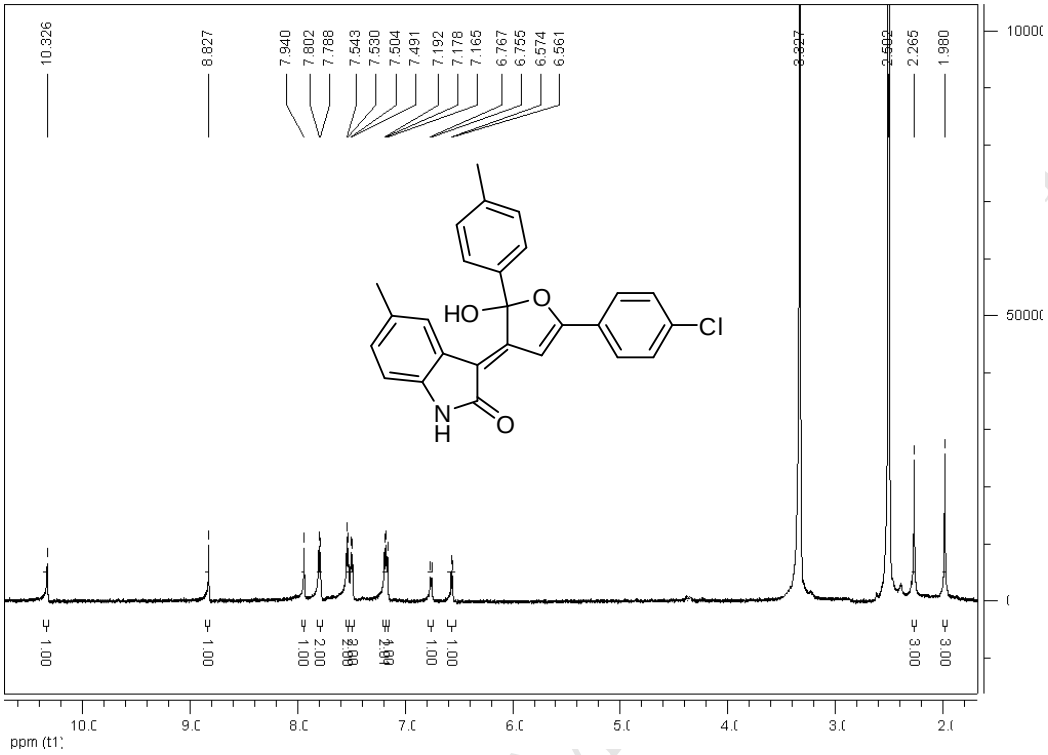


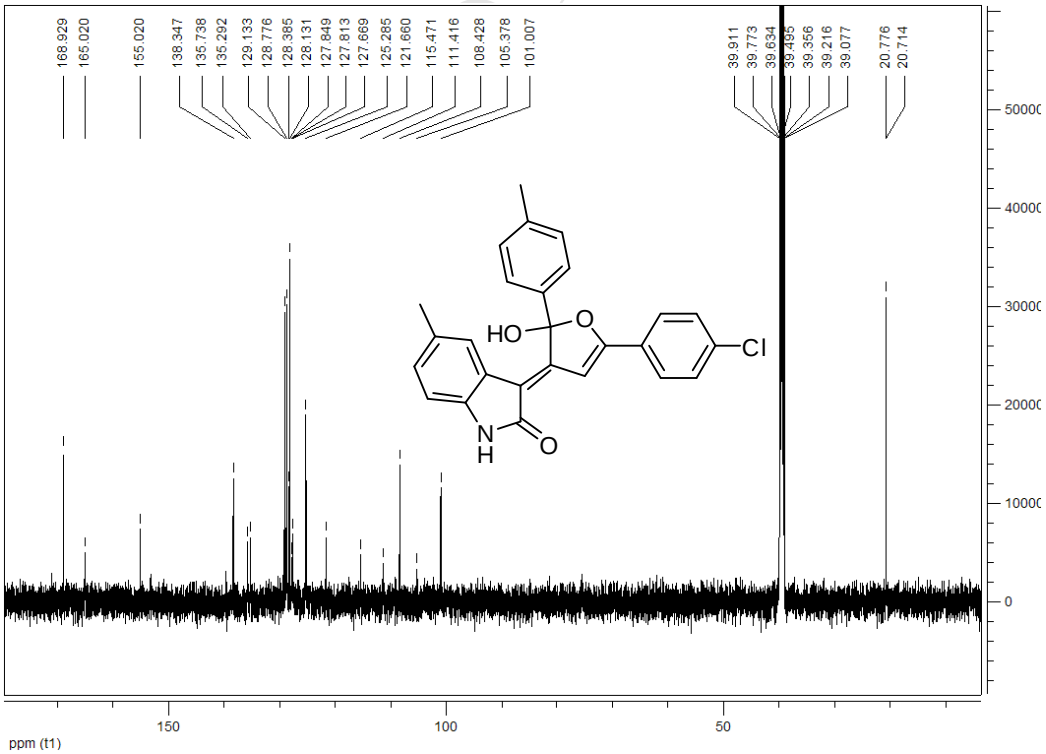
2-(4-chlorobenzoyl)-5'-methyl-3-(4-methylbenzoyl)spiro[cyclopropane-1,3'- indolin]-2'-one (4m):



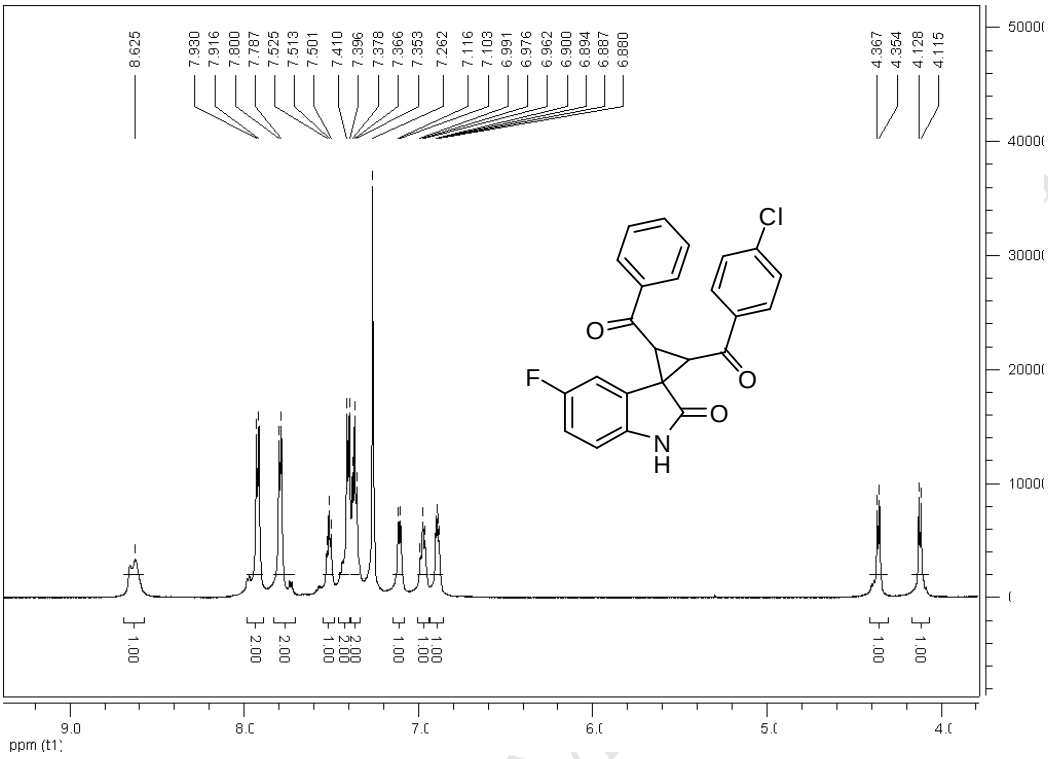


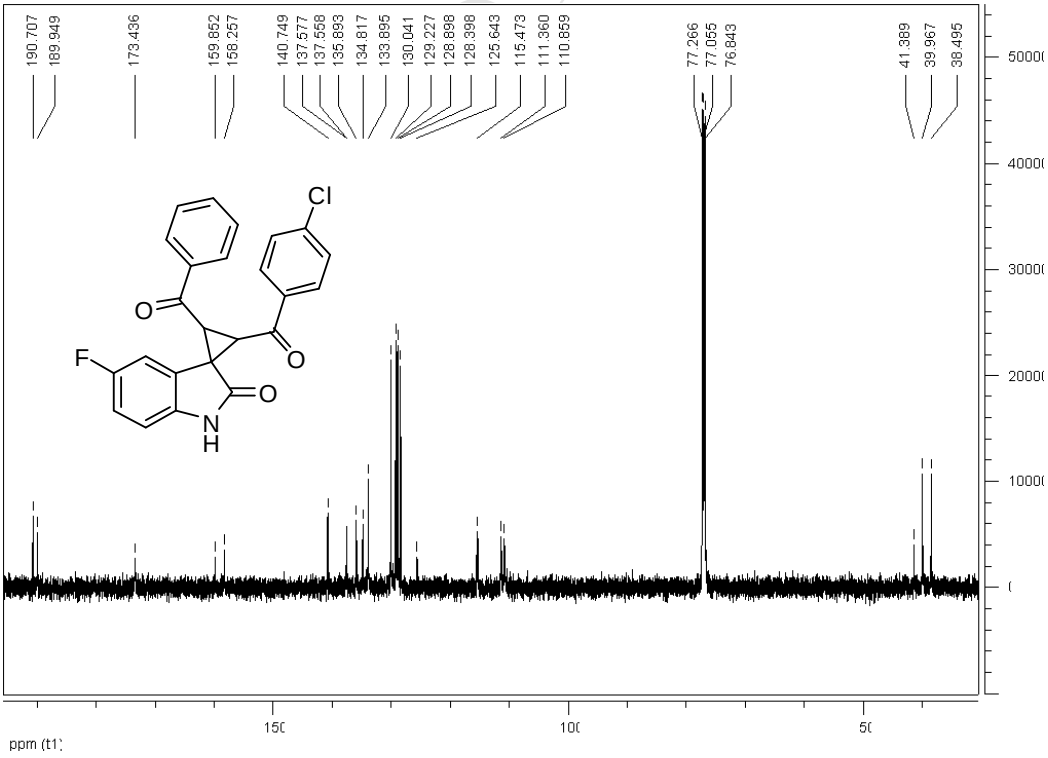
3-(5-(4-chlorophenyl)-2-hydroxy-2-p-tolylfuran-3(2H)-ylidene)-5-methylindolin-2-one (5m):



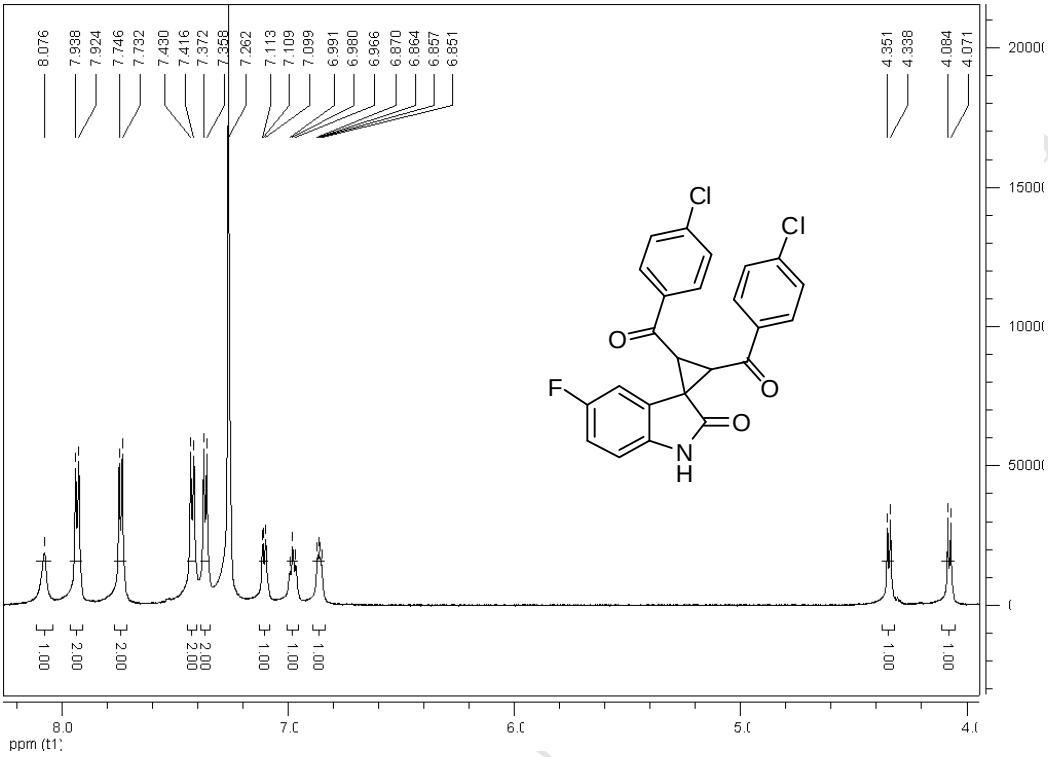


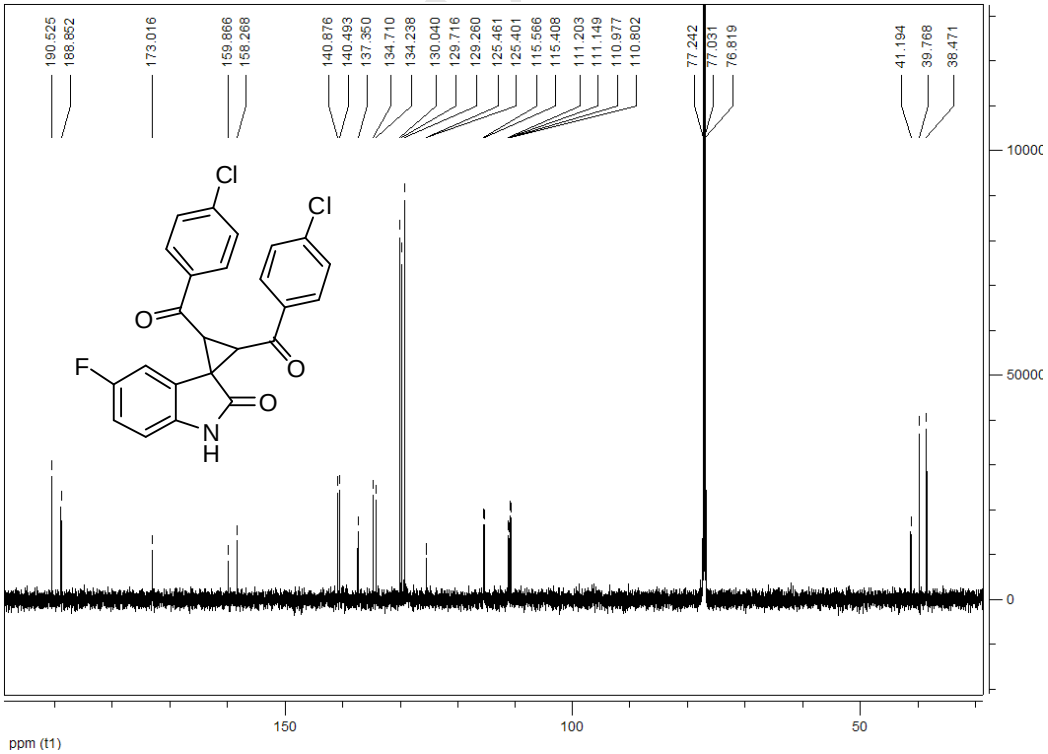
2-benzoyl-3-(4-chlorobenzoyl)-5'-fluorospiro[cyclopane-1,3'-indolin]-2'-one (4n):



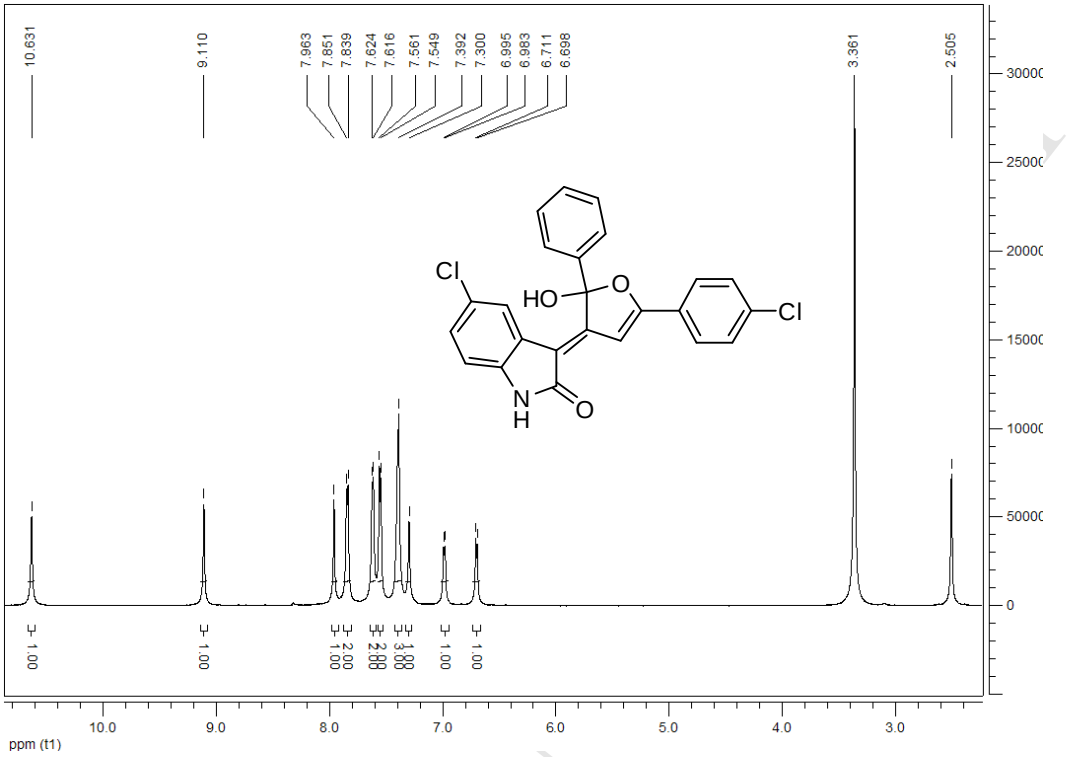


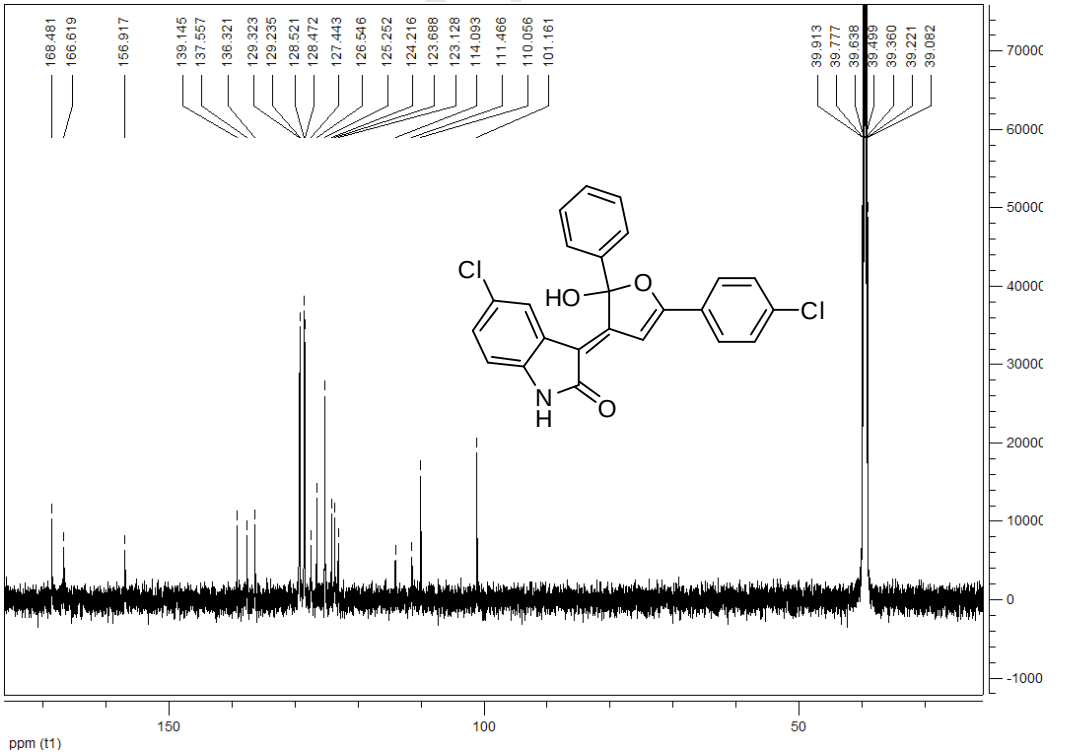
(5'-fluoro-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)-bis-((4-chlorophenyl)methanone) (4o):



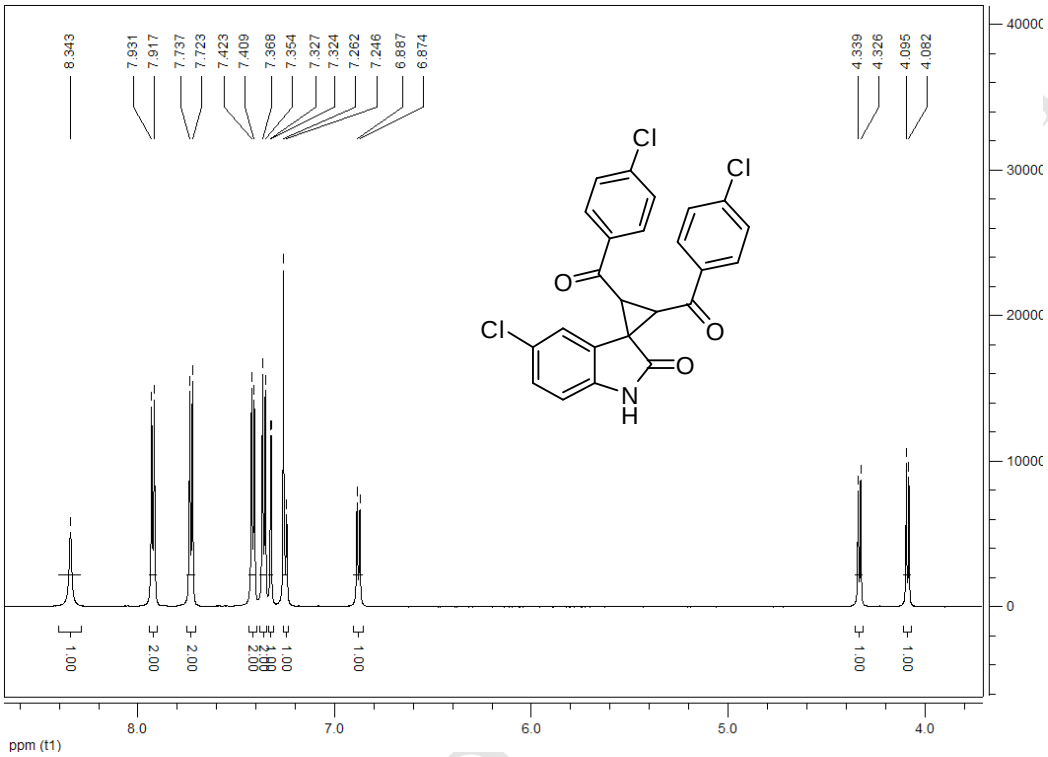


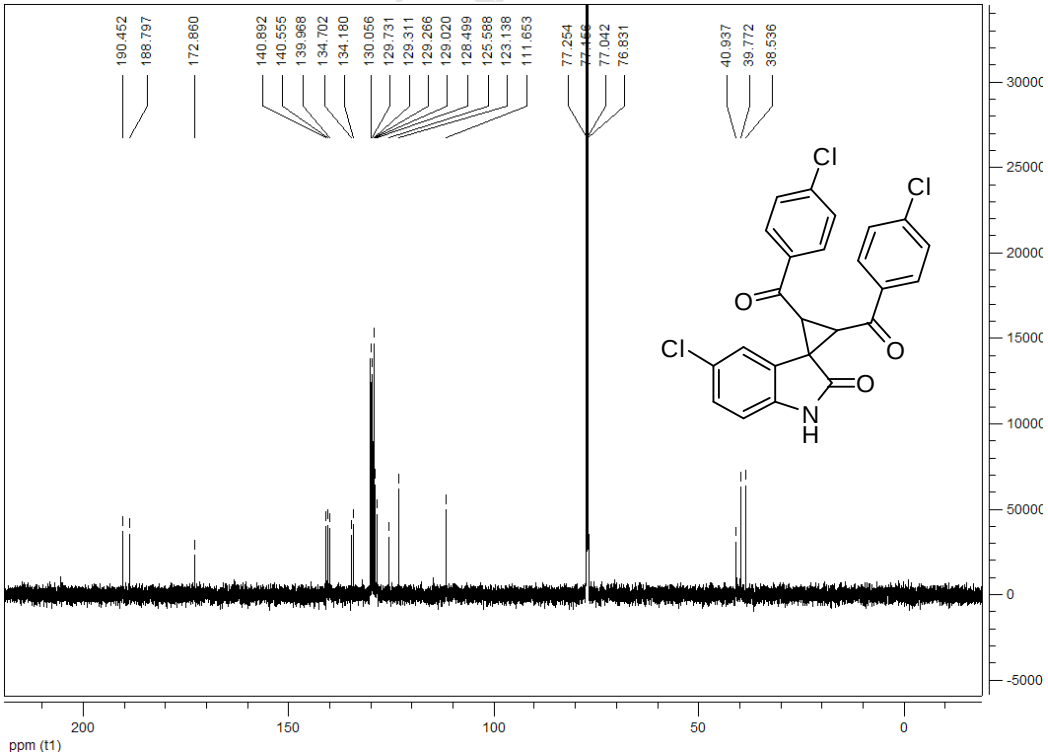
5-chloro-3-(5-(4-chlorophenyl)-2-hydroxy-2-phenylfuran-3(2H)-ylidene)indolin-2-one (5p):



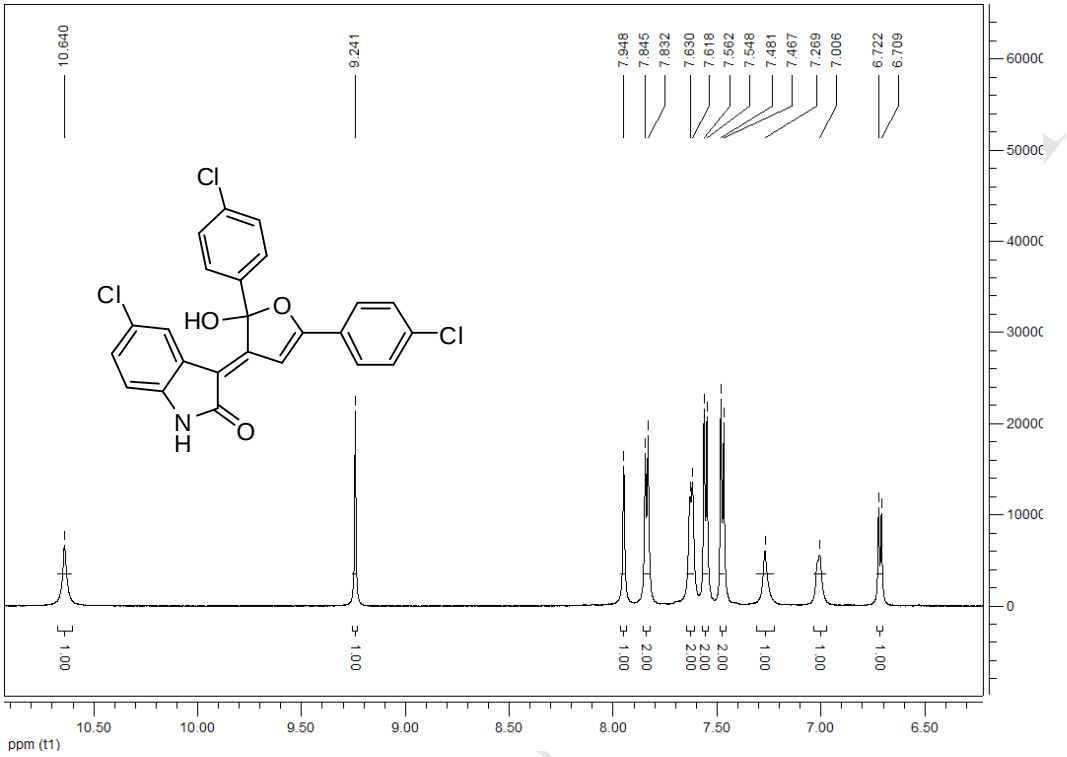


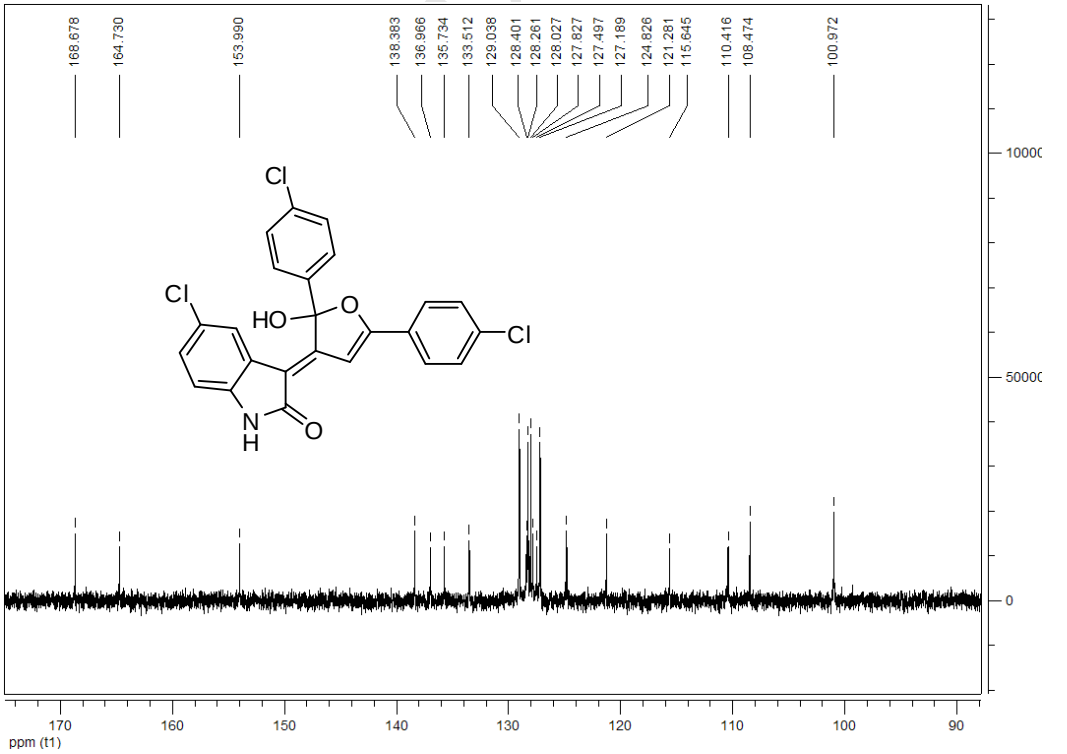
(5'-chloro-2'-oxospiro[cyclopropane-1,3'-indoline]-2,3-diyl)bis- ((4-chlorophenyl)methanone) (4q):



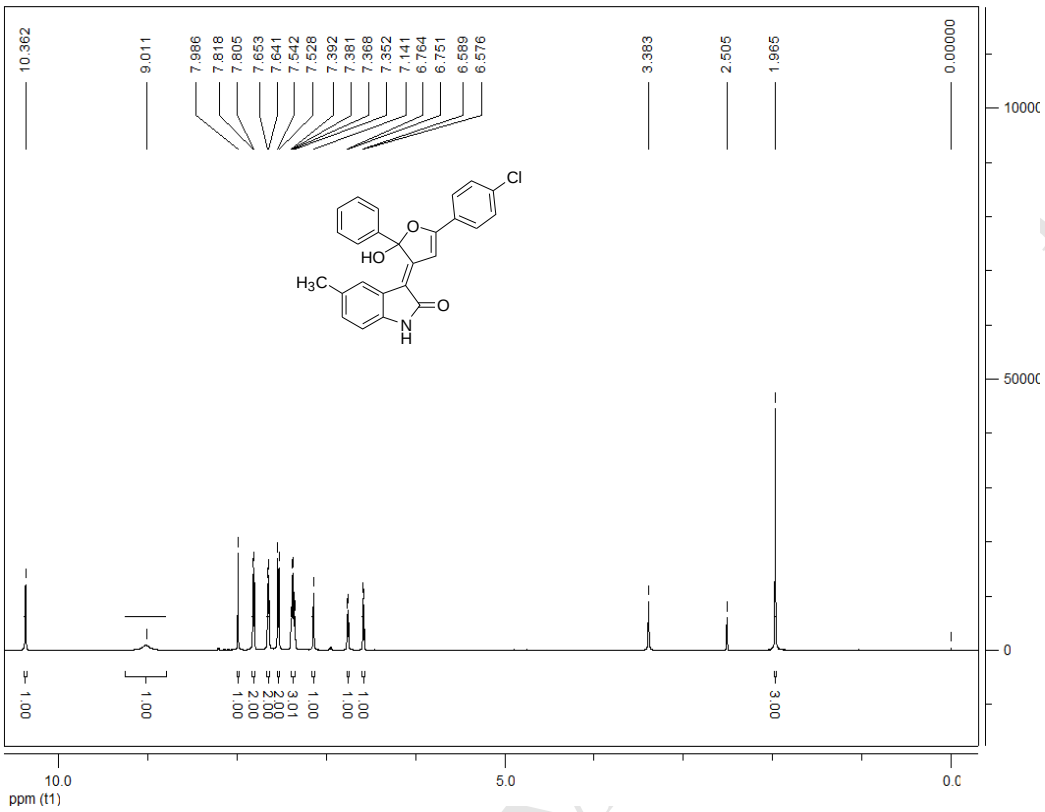


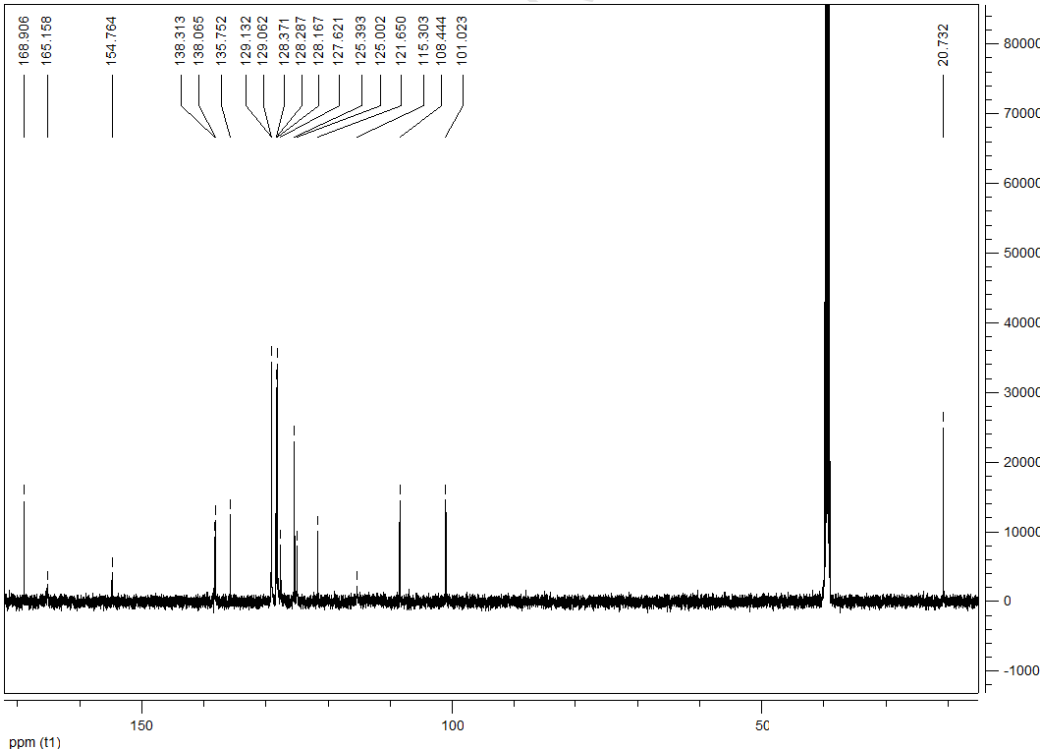
3-(2,5-bis(4-chlorophenyl)-2-hydroxyfuran-3(2H)-ylidene)-5-chloroindolin-2-one (5q):



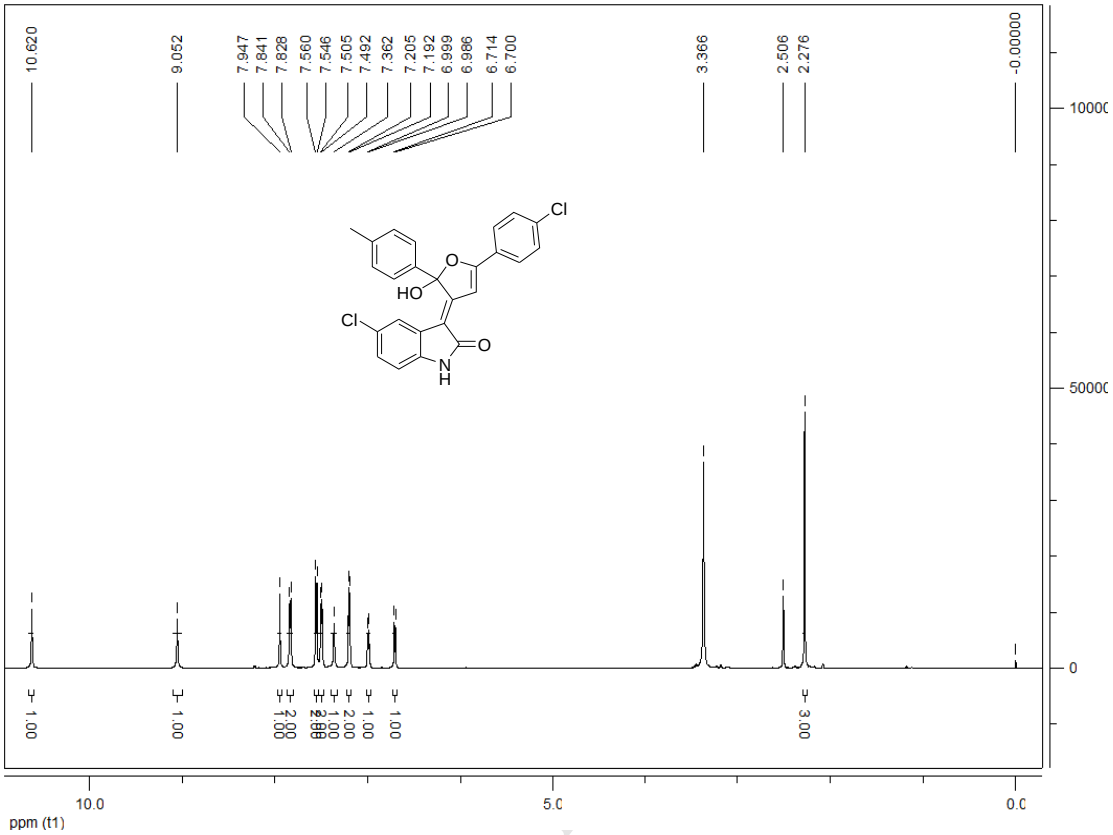


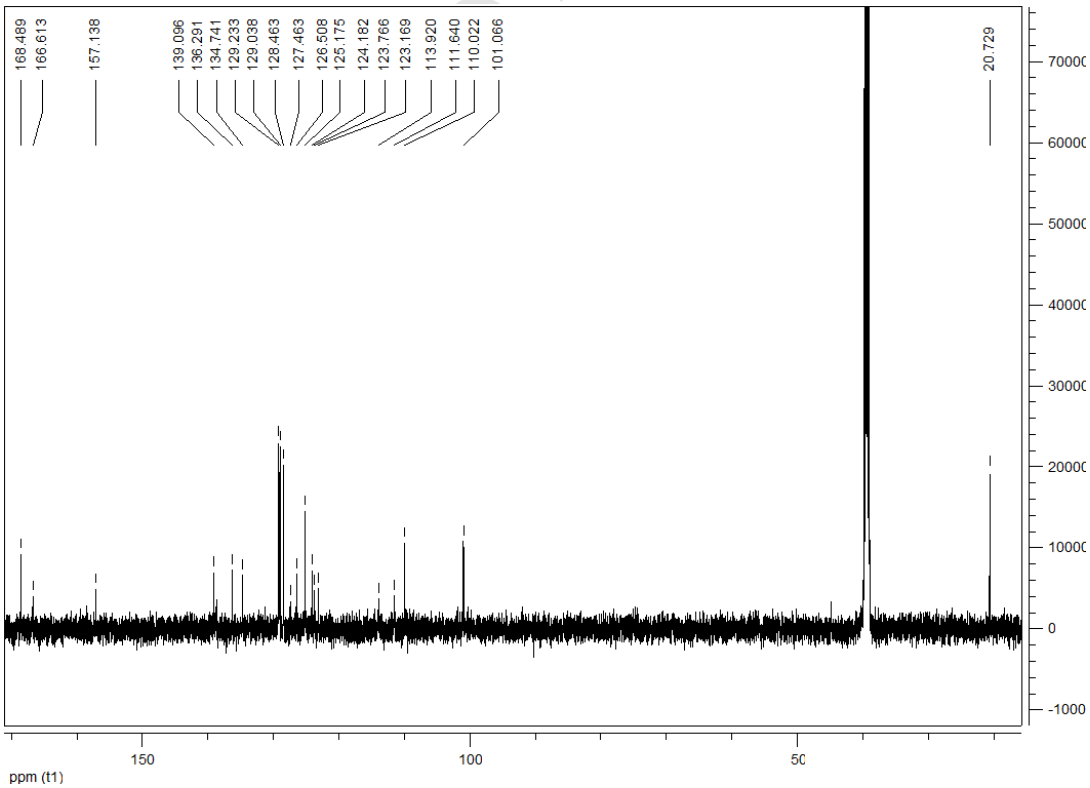
3-(5-(4-chlorophenyl)-2-hydroxy-2-phenylfuran-3(2H)-ylidene)-5-methylindolin-2-one (5r):





5-chloro-3-(5-(4-chlorophenyl)-2-hydroxy-2-*p*-tolylfuran-3(2H)-ylidene)indolin-2-one(5s):





3-(5-(4-chlorophenyl)-2-hydroxy-2-p-tolylfuran-3(2H)-ylidene)-5-fluoroindolin-2-one(5t):