Electronic Supplementary Material (ESI) for New Journal of Chemistry.


# DDQ dehydrogenative Diels-Alder reaction for efficient synthesis of functionalized spiro[carbazole-1,3'-indolines] and spiro[carbazole-1,5'-pyrimidines]

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## Supporting Information

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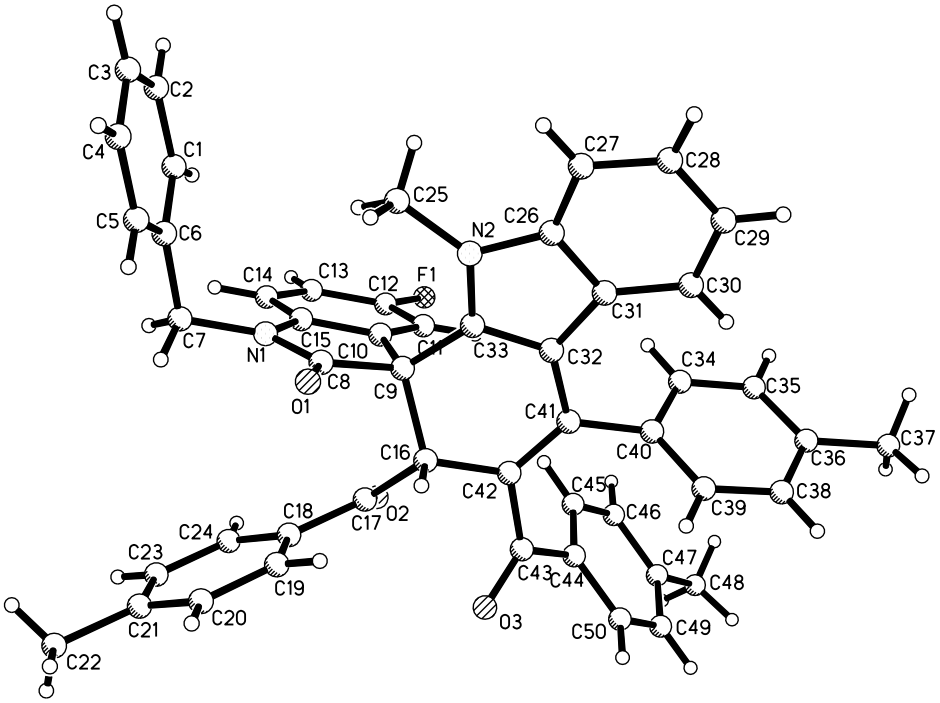


Fig. s1 Single crystal structure of the spiro compound **3a**

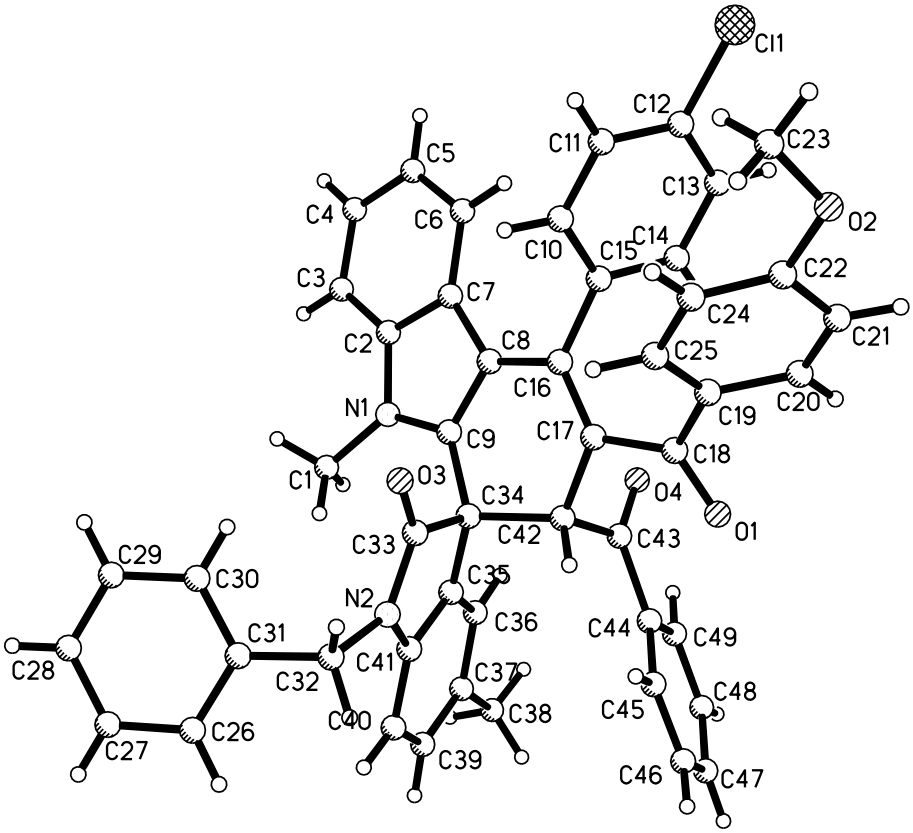


Fig. s2 Single crystal structure of the spiro compound **3b**

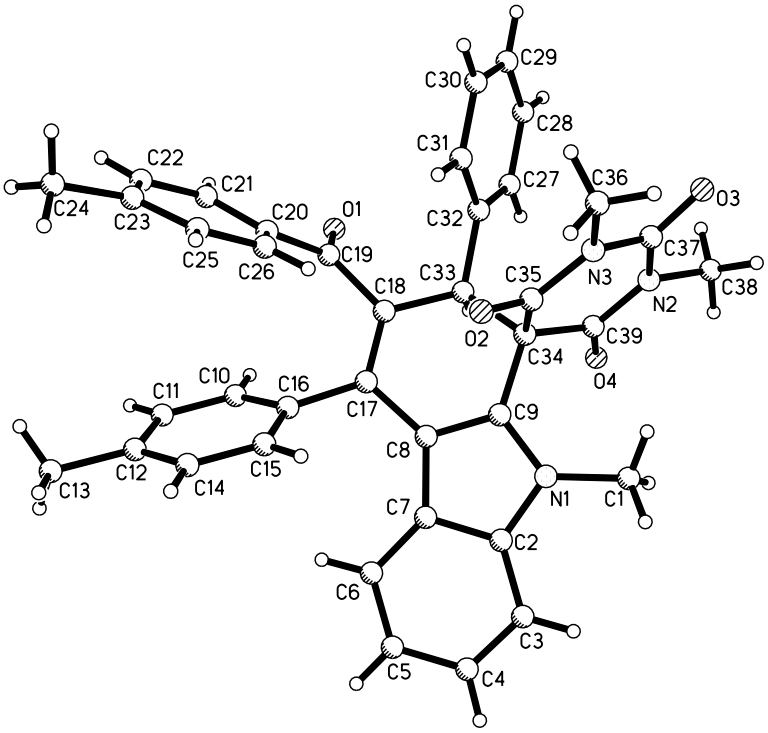


Fig. s3 Single crystal structure of the spiro compound **5b**

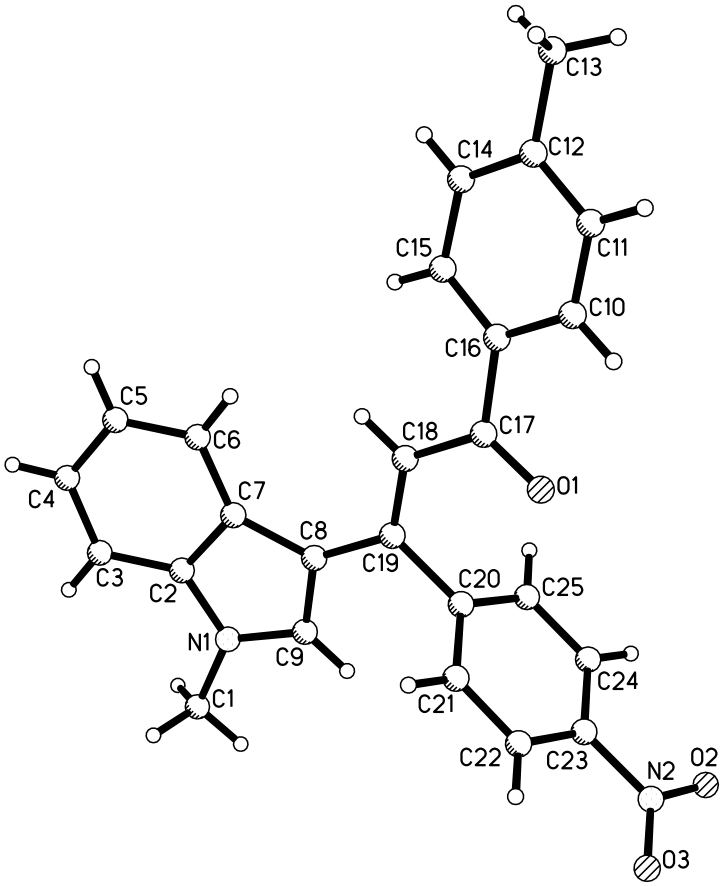


Fig. s4 Single crystal structure of the compound **A2**

**Table S1**The single crystal date of compounds **3a, 3b**

| Phase | **3a** | **3b** |
|---|---|---|
| Empirical formula | $C_{50}H_{39}FN_2O_3$ | $C_{49}H_{37}ClN_2O_4$ |
| Formula weight | 734.83 | 753.25 |
| Temperature/K | 296(2) K | 273(2) K |
| Wavelength/ Å | 0.71073 | 0.71073 |
| Crystal system | Triclinic | Triclinic |
| Space group | P-1 | P-1 |
| *a* /Å | 11.874(3) | 12.3059(11) |
| *b* / Å | 13.417(3) | 12.9944(10) |
| *c* / Å | 14.271(3) | 13.5855(11) |
| α (°) | 62.751(6) | 99.021(2) |
| β (°) | 75.230(7) | 99.217(3) |
| γ ( °) | 82.876(6) | 110.717(3) |
| *V* (Å $^3$) | 1954.4(7) | 1951.4(3) |
| *Z* | 2 | 2 |
| Calculated density (g·cm$^{-3}$) | 1.249 | 1.282 |
| Absorption coefficient(mm$^{-1}$) | 0.081 | 0.147 |
| *F*(000) | 772 | 788 |
| θ range /(°) | 2.216 to 24.712 deg. | 1.960 to 25.996 |
| Limiting indices | -13<=h<=13, -15<=k<=15, -16<=l<=16 | -15<=h<=15, -16<=k<=16, -16<=l<=16 |
| Reflections collected/unique | 22269 / 6468 [R(int) = 0.1010] | 27540 / 7613 [R(int) = 0.0837] |
| Completeness to theta | 97.2 % | 99.3 % |
| Max. and min. transmission | 0.984 and 0.978 | 0.971 and 0.963 |
| Refinement method | Full-matrix least-squares on F^2 | Full-matrix least-squares on F^2 |
| Data/restraints/parameters | 6468 / 0 / 509 | 7613 / 0 / 508 |
| Goodness-of-fit on $F^2$ | 1.008 | 1.034 |
| Final *R* indices[I>2sigma(I)] | R1 = 0.0568, wR2 = 0.1273 | R1 = 0.0749, wR2 = 0.1648 |
| *R* indices (all data) | R1 = 0.1620, wR2 = 0.1796 | R1 = 0.1978, wR2 = 0.2031 |
| Largest diff. peak and hole /(e ·Å$^{-3}$) | 0.340 and -0.369 | 0.448 and -0.270 |

**Table S2**The single crystal date of compounds **5b, A2**

| Phase | **5b** | **A2** |
|---|---|---|
| Empirical formula | $C_{39}H_{33}lN_3O_4$ | $C_{25}H_{20}N_2O_3$ |
| Formula weight | 607.68 | 396.43 |
| Temperature/K | 296(2) K | 296(2) K |
| Wavelength/ Å | 0.71073 | 0.71073 |
| Crystal system | Monoclinic | Monoclinic |
| Space group | P2(1)/c | P2(1)/c |
| *a* /Å | 12.362(4) | 17.8914(19) |
| *b* / Å | 14.904(4) | 5.5286(6) |
| *c* / Å | 18.652(5) | 20.497(2) |
| α (°) | 90 | 90 |
| β (°) | 108.489(9) | 92.686(3) |
| γ ( °) | 90 | 90 |
| *V* (Å $^3$) | 3259.3(16) | 2025.2(4) |
| *Z* | 4 | 4 |
| Calculated density (g·cm$^{-3}$) | 1.238 | 1.300 |
| Absorption coefficient(mm$^{-1}$) | 0.081 | 0.086 |
| *F*(000) | 1280 | 832 |
| θ range /(°) | 2.210 to 25.999 | 2.339 to 26.000 |
| Limiting indices | -10<=h<=15, -18<=k<=17, -23<=l<=19 | -21<=h<=22, -6<=k<=6, -25<=l<=25 |
| Reflections collected/unique | 15073 / 6274 [R(int) = 0.0412] | 20401 / 3920 [R(int) = 0.1054] |
| Completeness to theta | 97.6 % | 98.7 % |
| Max. and min. transmission | 0.7455 and 0.6531 | 0.7456 and 0.6594 |
| Refinement method | Full-matrix least-squares on F^2 | Full-matrix least-squares on F^2 |
| Data/restraints/parameters | 6274 / 48 / 421 | 3920 / 0 / 274 |
| Goodness-of-fit on $F^2$ | 1.011 | 1.018 |
| Final *R* indices[I>2sigma(I)] | R1 = 0.0558, wR2 = 0.1121 | R1 = 0.0672, wR2 = 0.1172 |
| *R* indices (all data) | R1 = 0.1228, wR2 = 0.1384 | R1 = 0.1938, wR2 = 0.1544 |
| Largest diff. peak and hole /(e ·Å$^{-3}$) | 0.191 and -0.131 | 0.193 and -0.164 |

**Computational Details**

All calculations here were performed with the Gaussian 16 package of the program.[1] All structures are subjected to the following optimizations at the M06-2X[2] level using the Def2-SVP[3] basis set. Dispersion correction to the DFT energies was included following Grimme's DFT-D3 scheme.[4] The local minimum structures and the transition state (TS) structures on the potential energy surface are confirmed by vibrational frequency analysis. Intrinsic reaction coordinate (IRC)[5] calculations were further conducted to verify the reaction pathways of the determined TS. Calculate the Gibbs free energy at 351.69 K (the refluxing temperature of ethanol) and a pressure 1 atm.

**Optimized Cartesian coordinates, total energies and the lowest vibrational frequencies obtained from DFT calculations**

All distances are in Å, energies in Hartree, and frequencies in $cm^{-1}$.

(1) Reactant complex

E (RM062X) = –1914.668653

Freq = 13.39

| | | | |
|---|---|---|---|
| C | 0.68918300 | -1.08241300 | -4.50942900 |
| H | 1.63744900 | -0.56850700 | -4.31215500 |
| H | 0.36398300 | -0.87063000 | -5.53970400 |
| C | 0.00959400 | 0.37705100 | -2.61460400 |
| C | -1.24169300 | 0.53858700 | -1.77411600 |
| C | -2.23172900 | -0.37831700 | -2.35102300 |
| C | -3.56978000 | -0.66384600 | -2.08761000 |
| H | -4.06828900 | -0.18934500 | -1.24492700 |
| C | -4.25002600 | -1.56655200 | -2.91444400 |
| C | -3.59856600 | -2.18795700 | -3.97976900 |
| H | -4.14574700 | -2.88896800 | -4.61266000 |
| C | -2.24906000 | -1.93230000 | -4.25306100 |
| H | -1.73673900 | -2.41763300 | -5.08410800 |
| C | -1.58750400 | -1.02271300 | -3.43760400 |
| C | -1.17825500 | 1.41122600 | -0.75066500 |
| H | -0.24045600 | 1.96609800 | -0.67315400 |
| C | -2.17888600 | 1.55636500 | 0.33294800 |
| C | -2.04620600 | 2.71830000 | 1.26576800 |
| C | -1.18561700 | 3.79569500 | 1.02144300 |

| | | | |
|---|---|---|---|
| H | -0.56951600 | 3.82581800 | 0.12303300 |
| C | -1.11412100 | 4.85018000 | 1.92979000 |
| H | -0.44310900 | 5.68726900 | 1.73398100 |
| C | -1.88976100 | 4.82830900 | 3.08724600 |
| C | -2.74457900 | 3.75268600 | 3.34065300 |
| H | -3.34129200 | 3.73096100 | 4.25356600 |
| C | -2.82584600 | 2.70622300 | 2.43098700 |
| H | -3.48512100 | 1.85429000 | 2.60147800 |
| C | -2.05288600 | -3.81592800 | -1.16787200 |
| H | -2.83522900 | -3.32171800 | -0.58035000 |
| H | -1.93974000 | -4.85488500 | -0.82331900 |
| H | -2.35908500 | -3.81234800 | -2.22329700 |
| C | 0.35395500 | -3.35869200 | -1.68274000 |
| C | 0.59417200 | -4.33867600 | -2.65351200 |
| H | -0.19125300 | -5.02671600 | -2.96959600 |
| C | 1.87126800 | -4.39968900 | -3.19246200 |
| H | 2.10225100 | -5.15214900 | -3.94791200 |
| C | 2.87107800 | -3.49673600 | -2.78116300 |
| H | 3.86291200 | -3.56046100 | -3.23058800 |
| C | 2.62856100 | -2.52764200 | -1.81686800 |
| H | 3.42234400 | -1.84138400 | -1.52406300 |
| C | 1.34922700 | -2.45277900 | -1.22954100 |
| C | 0.71378700 | -1.61683200 | -0.22377400 |
| C | -0.60203500 | -2.05144700 | -0.15616300 |
| C | 3.58921400 | -1.23358200 | 1.11788400 |
| H | 3.14086400 | -2.01837100 | 1.73113600 |
| C | 4.97435200 | -1.15061000 | 0.98730800 |
| H | 5.61313700 | -1.86261600 | 1.51218600 |
| C | 5.53785800 | -0.16293600 | 0.18049400 |
| C | 4.71162300 | 0.74751200 | -0.47928400 |
| H | 5.14861900 | 1.52619900 | -1.10623100 |
| C | 3.32649100 | 0.67401800 | -0.34297900 |
| H | 2.67531100 | 1.37224000 | -0.86614300 |
| C | 2.75909800 | -0.32635700 | 0.45050400 |
| C | 1.28056700 | -0.52079600 | 0.56035400 |
| C | 0.51012500 | 0.20440300 | 1.41086300 |
| C | 0.97049000 | 1.37189100 | 2.19748100 |
| C | 0.32081700 | 1.61386400 | 3.53635300 |
| C | -0.55214400 | 0.70016100 | 4.13917500 |
| H | -0.78727300 | -0.24404200 | 3.64641100 |
| C | -1.11446800 | 0.98129900 | 5.38299700 |
| H | -1.79228000 | 0.26341300 | 5.84723500 |
| C | -0.80354000 | 2.17370200 | 6.03620800 |
| C | 0.07606000 | 3.08285300 | 5.44632700 |

| | | | |
|---|---|---|---|
| H | 0.32514900 | 4.01327500 | 5.95952700 |
| C | 0.63440900 | 2.80363700 | 4.20279400 |
| H | 1.32445000 | 3.49445300 | 3.71624600 |
| N | -0.27194100 | -0.58911100 | -3.55591600 |
| N | -0.81635700 | -3.08831700 | -1.00578900 |
| O | 1.06004100 | 0.95798500 | -2.48525900 |
| O | -3.04073800 | 0.71298300 | 0.50995400 |
| O | 1.81308600 | 2.14342600 | 1.78614500 |
| H | -1.24242300 | 2.39138600 | 7.01153400 |
| H | 6.62159400 | -0.09935200 | 0.06953800 |
| H | -1.82186000 | 5.64945700 | 3.80304900 |
| H | 0.83366500 | -2.16603200 | -4.38190400 |
| H | -5.30060200 | -1.78471500 | -2.72068800 |
| H | -0.53213000 | -0.08789100 | 1.55008100 |
| H | -1.42866600 | -1.66188200 | 0.43807700 |

(2) Transition state

E (RM062X) = –1914.640961

Freq = –377.89

| | | | |
|---|---|---|---|
| C | 18.15625700 | 14.57452700 | 7.89709000 |
| H | 19.16041300 | 14.97340400 | 8.08824400 |
| H | 17.76441500 | 15.00373800 | 6.96196800 |
| C | 17.80607100 | 15.64847500 | 10.11720500 |
| C | 16.70921100 | 15.72422300 | 11.08923100 |
| C | 15.54581600 | 15.12458600 | 10.45454300 |
| C | 14.20675500 | 14.91697800 | 10.80364600 |
| H | 13.83447300 | 15.25022500 | 11.77113100 |
| C | 13.35141300 | 14.27204500 | 9.90436500 |
| C | 13.80975100 | 13.82281900 | 8.66321700 |
| H | 13.12196800 | 13.32727700 | 7.97625400 |
| C | 15.14441500 | 14.00948100 | 8.28756800 |
| H | 15.51338600 | 13.66600200 | 7.32000300 |
| C | 15.99225700 | 14.64923700 | 9.18477200 |
| C | 17.00383600 | 16.48669900 | 12.25940800 |
| H | 17.72424400 | 17.28226000 | 12.05371300 |
| C | 15.86630900 | 16.80623600 | 13.18921500 |
| C | 15.75114100 | 18.17099300 | 13.79066900 |
| C | 16.52842300 | 19.25817600 | 13.37469900 |
| H | 17.23004800 | 19.15799700 | 12.54709100 |
| C | 16.41818800 | 20.48317400 | 14.03103300 |
| H | 17.02700500 | 21.32727200 | 13.70499400 |
| C | 15.53862800 | 20.62596400 | 15.10269700 |
| C | 14.75571900 | 19.54523500 | 15.51711400 |

| | | | |
|---|---|---|---|
| H | 14.07045300 | 19.65654700 | 16.35862600 |
| C | 14.85919000 | 18.32489300 | 14.86178100 |
| H | 14.27247400 | 17.45949800 | 15.17229700 |
| C | 15.69307300 | 11.72058600 | 10.89785500 |
| H | 14.94384600 | 12.14371200 | 11.57626900 |
| H | 15.79087100 | 10.64041700 | 11.08079900 |
| H | 15.35520600 | 11.89425300 | 9.86605300 |
| C | 18.14844200 | 12.06483100 | 10.47699400 |
| C | 18.34509500 | 11.16262800 | 9.43281600 |
| H | 17.52525000 | 10.56085800 | 9.03883200 |
| C | 19.63011900 | 11.07790900 | 8.90328000 |
| H | 19.82819800 | 10.38514700 | 8.08417100 |
| C | 20.66847100 | 11.88184800 | 9.39707500 |
| H | 21.65987700 | 11.80778700 | 8.94882500 |
| C | 20.46200900 | 12.77384500 | 10.44730700 |
| H | 21.27811700 | 13.39702000 | 10.81189400 |
| C | 19.18591000 | 12.85654800 | 11.01760300 |
| C | 18.57384200 | 13.68700500 | 12.05707700 |
| C | 17.19389700 | 13.35989100 | 12.02270300 |
| C | 21.33660200 | 13.98738500 | 13.43527800 |
| H | 20.83586000 | 13.12667700 | 13.88398800 |
| C | 22.72568500 | 14.08770400 | 13.47709100 |
| H | 23.31148300 | 13.30960800 | 13.96881800 |
| C | 23.35991100 | 15.17960900 | 12.88652900 |
| C | 22.59772800 | 16.16882300 | 12.26403600 |
| H | 23.09016400 | 17.02291900 | 11.79675600 |
| C | 21.20816400 | 16.07781600 | 12.22716900 |
| H | 20.60689100 | 16.83050500 | 11.72348700 |
| C | 20.56593100 | 14.97848500 | 12.81025500 |
| C | 19.09301400 | 14.77036900 | 12.77452000 |
| C | 18.13281700 | 15.58246000 | 13.47440800 |
| C | 18.60003200 | 16.70823300 | 14.35120000 |
| C | 17.97766700 | 16.88727100 | 15.70789700 |
| C | 17.17606200 | 15.92376500 | 16.33035100 |
| H | 17.01136600 | 14.94964600 | 15.86967200 |
| C | 16.57395500 | 16.20056800 | 17.55733100 |
| H | 15.94981400 | 15.44505500 | 18.03630500 |
| C | 16.76632400 | 17.43874700 | 18.16714900 |
| C | 17.57502500 | 18.40029700 | 17.55642200 |
| H | 17.72616800 | 19.37043700 | 18.03219900 |
| C | 18.17982100 | 18.12386200 | 16.33645700 |
| H | 18.80489100 | 18.86010600 | 15.82958300 |
| N | 17.33593300 | 14.93872400 | 9.02093000 |
| N | 16.96027800 | 12.37689800 | 11.13661000 |

| | | | |
|---|---|---|---|
| O | 18.93945200 | 16.08453300 | 10.24763700 |
| O | 15.10735700 | 15.91559800 | 13.52179000 |
| O | 19.37414500 | 17.53609200 | 13.92204200 |
| H | 16.28630200 | 17.65550300 | 19.12294400 |
| H | 24.44788600 | 15.26039200 | 12.91131000 |
| H | 15.46202000 | 21.58426200 | 15.61932900 |
| H | 18.21864900 | 13.47864300 | 7.79693200 |
| H | 12.30650800 | 14.12117900 | 10.17918400 |
| H | 17.33461000 | 14.98506600 | 13.91380400 |
| H | 16.38972700 | 13.70946400 | 12.66818000 |

(3) Product

E(RM062X) = -1914.686945

Freq = 15.95

| | | | |
|---|---|---|---|
| C | 16.13077900 | 16.22759000 | 8.15922700 |
| H | 17.16827300 | 16.47389300 | 7.90317400 |
| H | 15.50922700 | 17.13524900 | 8.09573000 |
| C | 17.25303100 | 15.67462200 | 10.30269900 |
| C | 16.83358600 | 15.14517600 | 11.68792000 |
| C | 15.33677600 | 14.99450600 | 11.52905900 |
| C | 14.35985000 | 14.58109100 | 12.41943600 |
| H | 14.61842700 | 14.32237500 | 13.44861500 |
| C | 13.02524300 | 14.54109200 | 11.99047500 |
| C | 12.69287400 | 14.90926900 | 10.68807600 |
| H | 11.64964800 | 14.88048300 | 10.37006400 |
| C | 13.67281300 | 15.31815500 | 9.77472600 |
| H | 13.40984700 | 15.60594800 | 8.75656700 |
| C | 14.98883800 | 15.34704600 | 10.21636800 |
| C | 17.31900000 | 16.14127900 | 12.79067700 |
| H | 18.02793600 | 16.83404200 | 12.31828500 |
| C | 16.12835100 | 16.93050200 | 13.32174600 |
| C | 15.45799200 | 17.88055000 | 12.37367900 |
| C | 16.10910900 | 18.42412500 | 11.26023300 |
| H | 17.16787800 | 18.23004000 | 11.08016100 |
| C | 15.40417400 | 19.23313600 | 10.36882400 |
| H | 15.91818200 | 19.66587500 | 9.50895300 |
| C | 14.04989300 | 19.49002400 | 10.57820400 |
| C | 13.40052100 | 18.95802000 | 11.69487500 |
| H | 12.34175000 | 19.16207800 | 11.86033300 |
| C | 14.10459600 | 18.16488700 | 12.59357700 |
| H | 13.62298000 | 17.73015700 | 13.47079300 |
| C | 16.37236700 | 12.28094600 | 10.12285400 |
| H | 15.51469000 | 12.38306300 | 10.80119200 |

| | | | |
|---|---|---|---|
| H | 16.56829400 | 11.20636200 | 9.97272200 |
| H | 16.08510300 | 12.71940700 | 9.15127300 |
| C | 18.82120500 | 12.54923800 | 10.40862000 |
| C | 19.25946700 | 11.75190900 | 9.34987000 |
| H | 18.55635800 | 11.34451000 | 8.62308000 |
| C | 20.63018300 | 11.50946300 | 9.23645200 |
| H | 20.99208700 | 10.89029800 | 8.41360700 |
| C | 21.54360800 | 12.04978100 | 10.14385700 |
| H | 22.60927200 | 11.85385000 | 10.02470300 |
| C | 21.09884500 | 12.84361800 | 11.20480000 |
| H | 21.81218100 | 13.26725000 | 11.91167200 |
| C | 19.73439200 | 13.08450200 | 11.34732000 |
| C | 18.94568400 | 13.86573600 | 12.30450700 |
| C | 17.49417000 | 13.74165300 | 11.89260000 |
| C | 21.32734400 | 13.80717300 | 14.45235300 |
| H | 20.82430200 | 12.84359600 | 14.56167700 |
| C | 22.62388200 | 13.98159700 | 14.93026000 |
| H | 23.13769600 | 13.15318700 | 15.42039500 |
| C | 23.26049000 | 15.21476800 | 14.78797500 |
| C | 22.59599200 | 16.26811300 | 14.16140300 |
| H | 23.09037700 | 17.23316500 | 14.04137300 |
| C | 21.29843700 | 16.09414600 | 13.68268900 |
| H | 20.78066000 | 16.92070200 | 13.19288200 |
| C | 20.64949400 | 14.86056000 | 13.82460300 |
| C | 19.25824600 | 14.66847100 | 13.33817200 |
| C | 18.08845900 | 15.45125000 | 13.92892800 |
| C | 18.54729000 | 16.49643600 | 14.94565900 |
| C | 18.96591100 | 16.03605300 | 16.30825100 |
| C | 18.93618900 | 14.69192500 | 16.69575100 |
| H | 18.60334700 | 13.92225300 | 15.99748100 |
| C | 19.35320800 | 14.32270300 | 17.97259800 |
| H | 19.33141200 | 13.27319000 | 18.26889000 |
| C | 19.79994300 | 15.29374000 | 18.86796500 |
| C | 19.83252200 | 16.63634400 | 18.48625300 |
| H | 20.18360800 | 17.39519400 | 19.18687300 |
| C | 19.41832400 | 17.00515000 | 17.21107400 |
| H | 19.43442300 | 18.04513600 | 16.88316700 |
| N | 16.12953400 | 15.71175800 | 9.50273800 |
| N | 17.52594500 | 12.92383900 | 10.69847700 |
| O | 18.35901200 | 16.02571900 | 9.97857200 |
| O | 15.67665100 | 16.71453000 | 14.42134700 |
| O | 18.59434200 | 17.66537200 | 14.64087300 |
| H | 20.12612500 | 15.00275400 | 19.86784900 |
| H | 24.27448900 | 15.35426400 | 15.16561500 |

| | | | |
|---|---|---|---|
| H | 13.49856500 | 20.11410200 | 9.87285300 |
| H | 15.74769000 | 15.47856000 | 7.44996900 |
| H | 12.24338200 | 14.22891700 | 12.68311600 |
| H | 17.39488000 | 14.76752100 | 14.44858400 |
| H | 16.91143900 | 13.24688200 | 12.69576300 |

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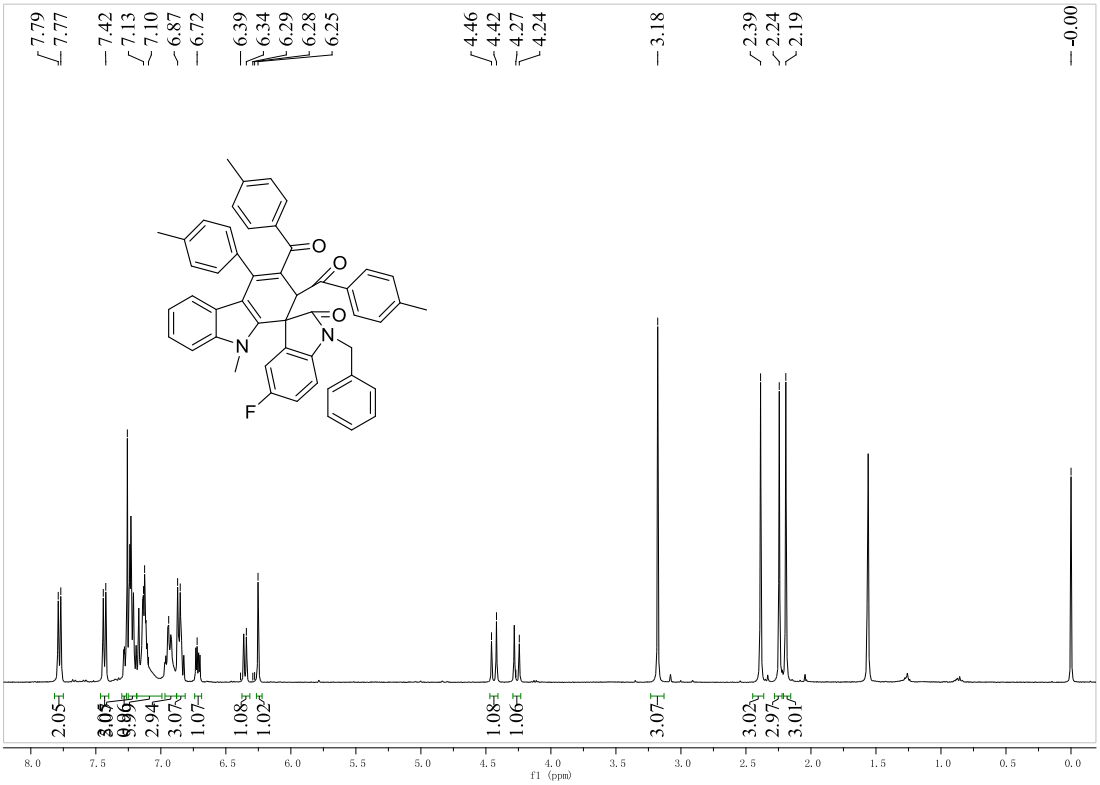
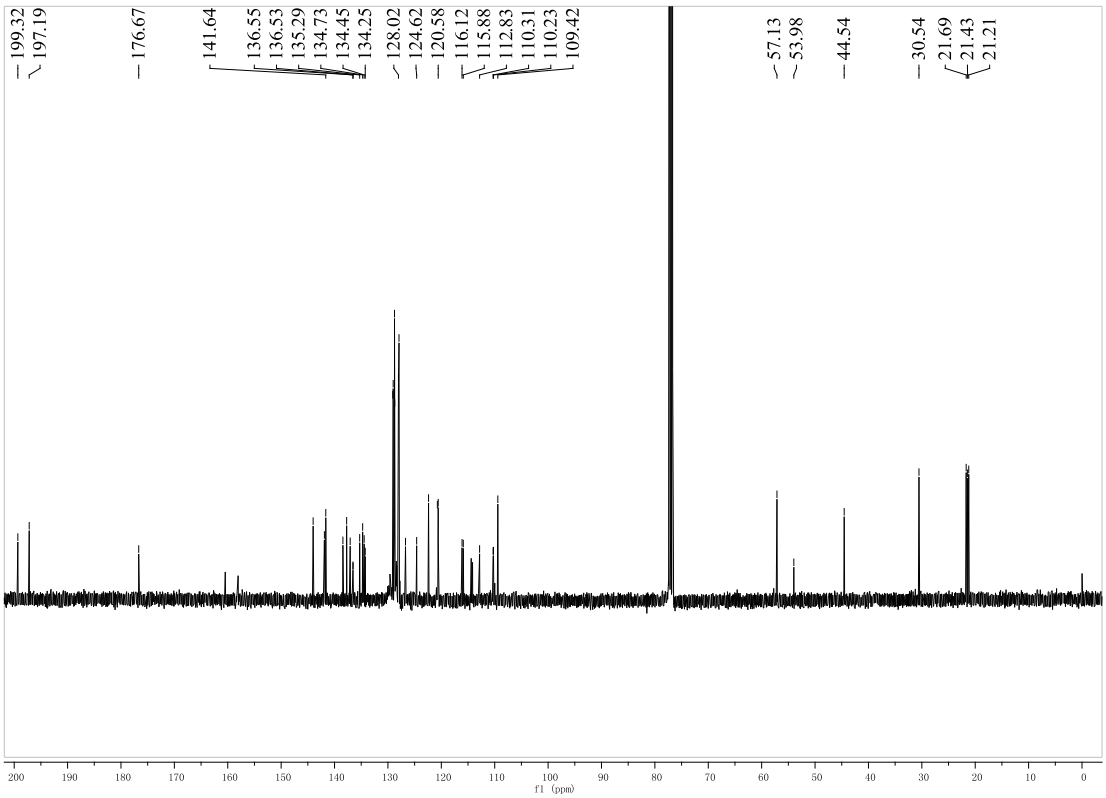
**(1'-Benzyl-5'-fluoro-9-methyl-2'-oxo-4-(*p*-tolyl)-2,9-dihydrospiro[carbazole-1,3'-indoline]-2,3'-diyl)bis(*p*-tolylmethanone) (3a):**

Yellow solid, 302 mg, 80%, m.p. 171-173 °C; $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.80 (d, *J* = 8.0 Hz, 2H, ArH), 7.43 (d, *J* = 8.0 Hz, 2H, ArH), 7.28-7.26 (m, 1H, ArH), 7.26-7.21 (m, 5H, ArH), 7.19-7.11 (m, 6H, ArH), 6.97-6.92 (m, 3H, ArH), 6.87-6.82 (m, 3H, ArH), 6.73-6.69 (m, 1H, ArH), 6.35 (d, *J* = 8.0 Hz, 1H, ArH), 6.25 (s, 1H, CH), 4.43 (d, *J* = 16.0 Hz, 1H, CH), 4.26 (d, *J* = 16.0 Hz, 1H, CH), 3.18 (s, 3H, $CH_3$), 2.39 (s, 3H, $CH_3$), 2.24 (s, 3H, $CH_3$), 2.19 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 199.3, 197.1, 176.6, 144.0, 141.8, 141.6, 138.4, 137.7, 137.0, 136.5, 135.2, 134.7, 134.4, 129.0, 128.7, 128.0, 127.9, 126.7, 124.6, 122.3, 120.7, 115.8, 112.8, 110.3, 109.4, 57.1, 53.9, 44.5, 30.5, 21.6, 21.4, 21.2; IR (KBr) υ: 3082, 3064, 2918, 1720, 1605, 1482, 1320, 1267, 1172, 1009, 952, 805, 743, 612, 477 cm$^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{50}H_{39}FN_2O_3$ ([M+Na]$^+$): 757.2837, found: 757.2832.

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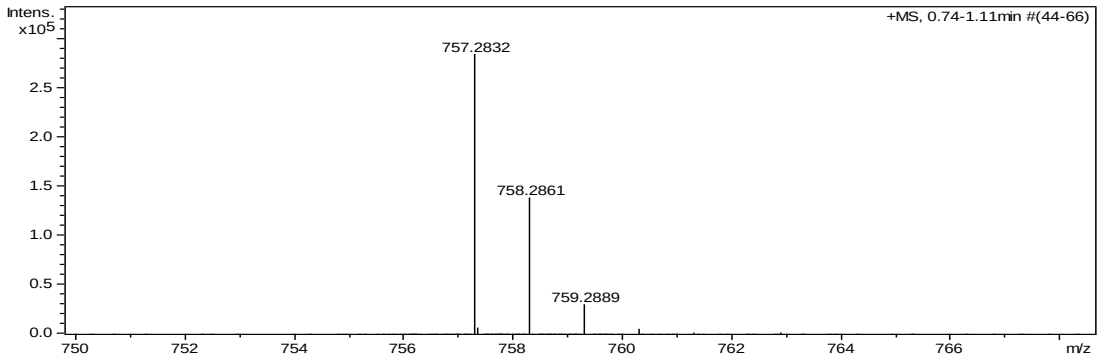
197.19
176.67
141.64
136.55
136.53
135.29
134.73
134.45
134.25
128.02
124.62
120.58
116.12
115.88
112.83
110.31
110.23
109.42
57.13
53.98
44.54
30.54
21.69
21.43
21.21
200 190 180 170 160 150 140 130 120 110 100 90 80 70 60 50 40 30 20 10 0
f1 (ppm)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x10^5
+MS, 0.74-1.11min #(44-66)
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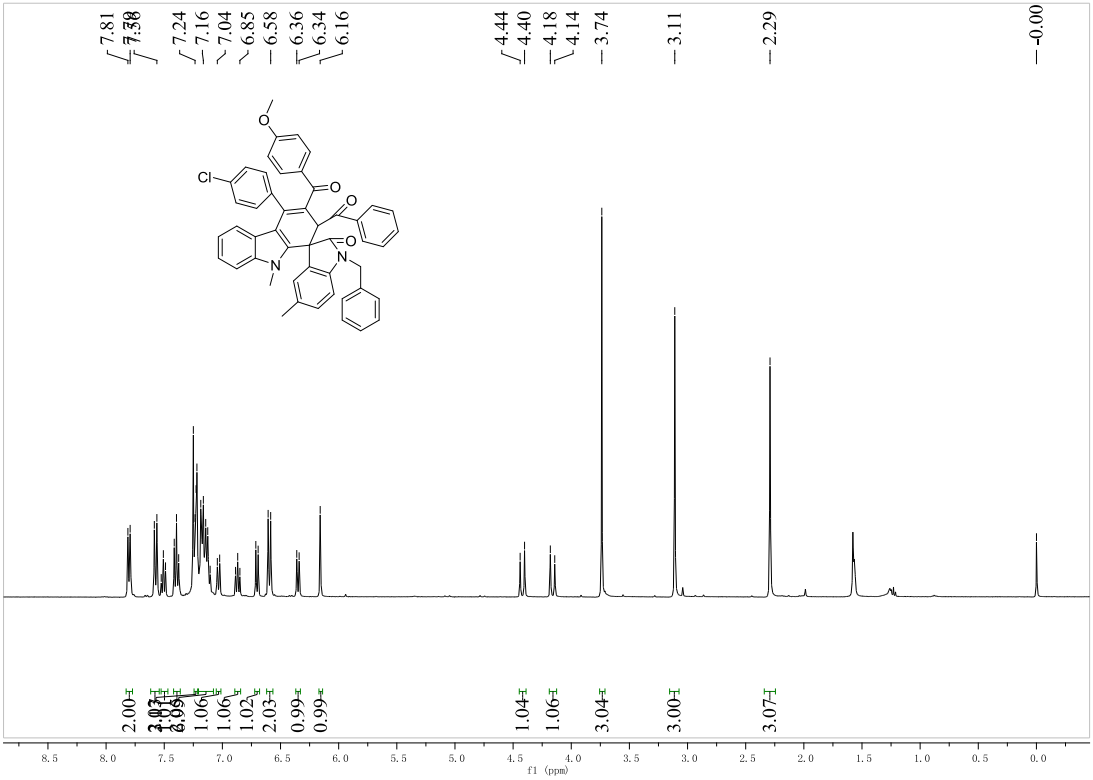
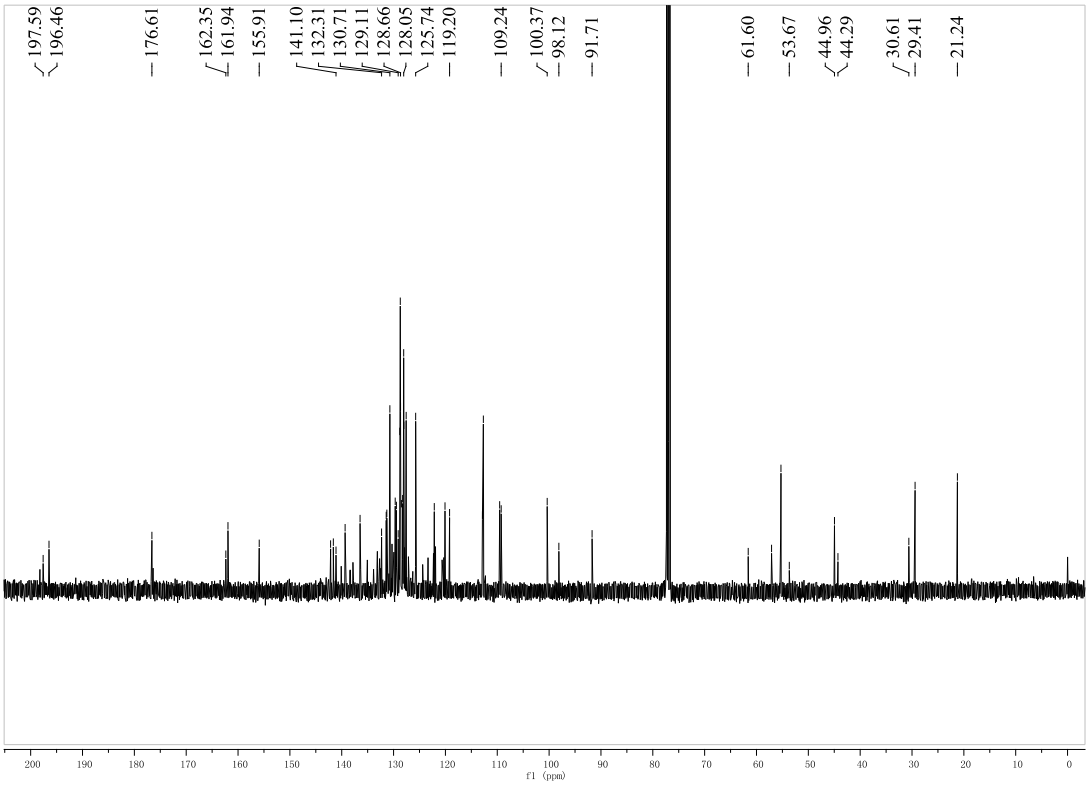
758.2861
759.2889
2.5
2.0
1.5
1.0
0.5
0.0
750 752 754 756 758 760 762 764 766
m/z

**2-Benzoyl-1'-benzyl-4-(4-chlorophenyl)-3-(4-methoxybenzoyl)-5',9-dimethyl-2,9-dihydrospiro[carbazole-1,3'-indolin]-2'-one (3b):**

Yellow solid, 302 mg, 78%, m.p. 159-162 $^{o}$C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.80 (d, *J* = 8.0 Hz, 2H, ArH), 7.57 (d, *J* = 8.0 Hz, 2H, ArH), 7.53-7.49 (m, 1H, ArH), 7.41-7.38 (m, 2H, ArH), 7.25-7.22 (m, 5H, ArH), 7.19-7.11 (m, 7H, ArH), 7.03 (d, *J* = 8.0 Hz, 1H, ArH), 6.89-6.85 (m, 1H, ArH), 6.70 (d, *J* = 8.0 Hz, 1H, ArH), 6.69 (d, *J* = 8.0 Hz, 2H, ArH), 6.35 (d, *J* = 8.0 Hz, 1H, ArH), 6.16 (s, 1H, CH), 4.42 (d, *J* = 16.0 Hz, 1H, CH), 4.16 (d, *J* = 16.0 Hz, 1H, CH), 3.74 (s, 3H, $OCH_3$), 3.11 (s, 3H, $CH_3$), 2.29 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 197.5, 196.4, 176.6, 162.3, 161.9, 155.9, 142.1, 141.6, 139.3, 129.6, 128.8, 128.7, 125.7, 122.1, 120.0, 119.1, 112.8, 112.6, 109.5, 109.2, 100.3, 61.5, 57.0, 53.6, 44.9, 29.4, 21.2; IR (KBr) υ: 3112, 3061, 2918, 1711, 1605, 1501, 1455, 1412, 1325, 1257, 1177, 1124, 1086, 1025, 959, 822, 769, 746, 698, 606, 478 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{49}H_{37}ClN_2O_4$ ($[M+Na]^+$): 775.2334, found: 775.2315.

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162.35
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155.91
141.10
132.31
130.71
129.11
128.66
128.05
125.74
119.20
109.24
100.37
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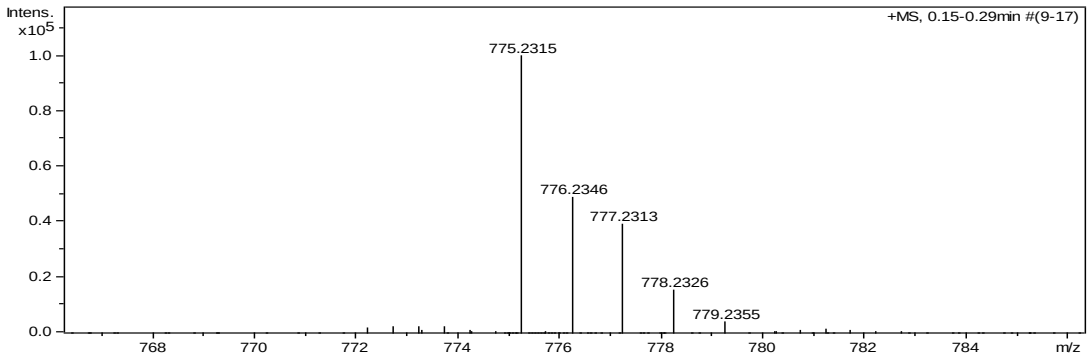
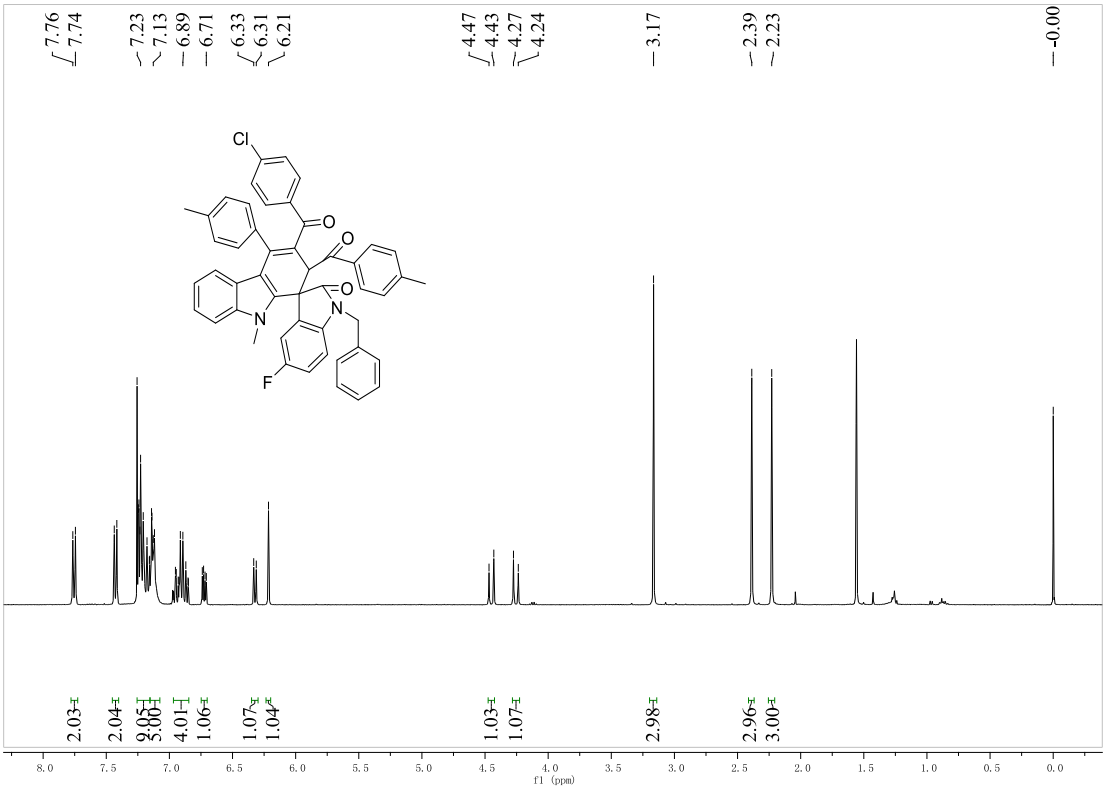
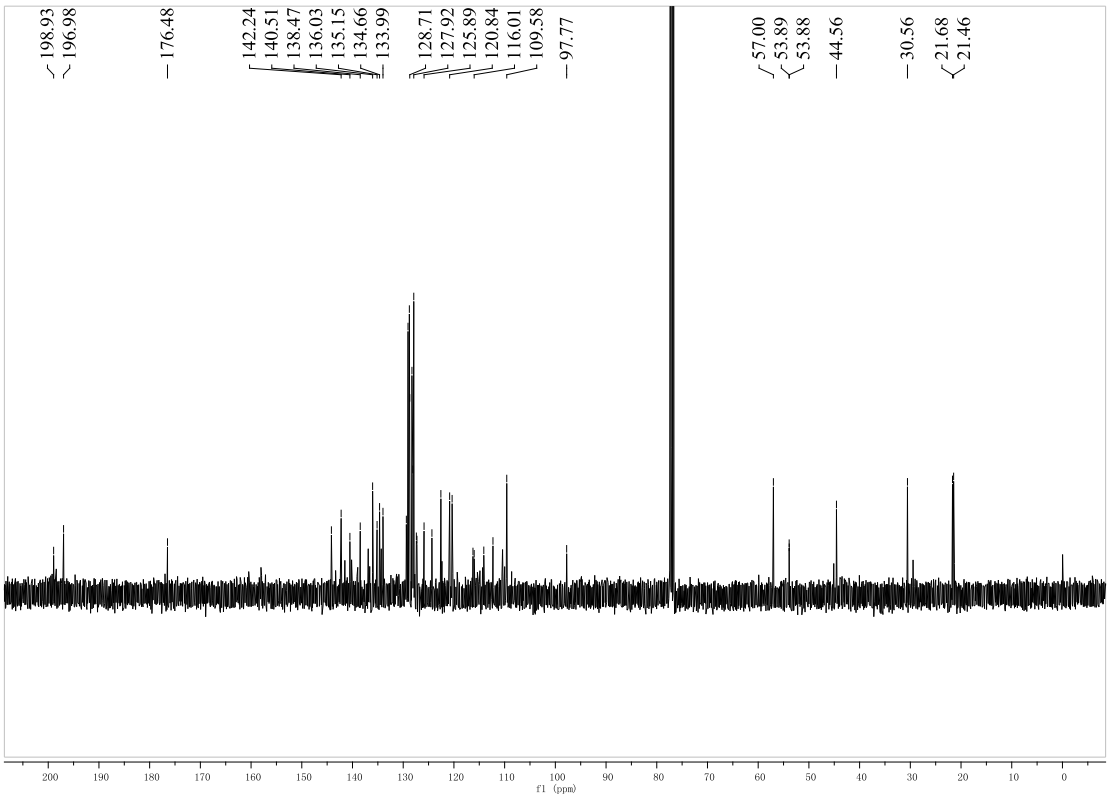
91.71
61.60
53.67
44.96
44.29
30.61
29.41
21.24
f1 (ppm)


Intens.
x10^5
+MS, 0.15-0.29min #(9-17)
775.2315
776.2346
777.2313
778.2326
779.2355
m/z

**1'-Benzyl-3-(4-chlorobenzoyl)-5'-fluoro-9-methyl-2-(4-methylbenzoyl)-4-(*p*-tolyl)-2,9-dihydro spiro[carbazole-1,3'-indolin]-2'-one (3c):**

Yellow solid, 292 mg, 75%, m.p. 173-176 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.76 (d, *J* = 8.0 Hz, 2H, ArH), 7.45 (d, *J* = 8.0 Hz, 2H, ArH), 7.26-7.18 (m, 9H, ArH), 7.16-7.12 (m, 5H, ArH), 6.95-6.85 (m, 4H, ArH), 6.74-6.71 (m, 1H, ArH), 6.32 (d, *J* = 8.0 Hz, 1H, ArH), 6.22 (s, 1H, CH), 4.45 (d, *J* = 15.2 Hz, 1H, CH), 4.26 (d, *J* = 15.2 Hz, 1H, CH), 3.17 (s, 3H, $CH_3$), 2.39 (s, 3H, $CH_3$), 2.23 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 198.9, 196.9, 176.4, 144.1, 142.2, 140.5, 138.4, 136.0, 135.1, 134.6, 133.9, 129.3, 129.0, 128.7, 128.2, 128.1, 127.9, 125.8, 124.3, 122.5, 120.8, 120.3, 116.2, 116.0, 114.1, 112.3, 109.5, 97.7, 57.0, 53.8, 44.5, 30.5, 21.6, 21.4; IR (KBr) υ: 3071, 3063, 2960, 2028, 1712, 1604, 1481, 1321, 1165, 1089, 956, 847, 700, 603, 480 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{49}H_{36}ClFN_2O_3$ ($[M+Na]^+$): 779.2291, found: 779.2075.

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136.03
135.15
134.66
133.99
128.71
127.92
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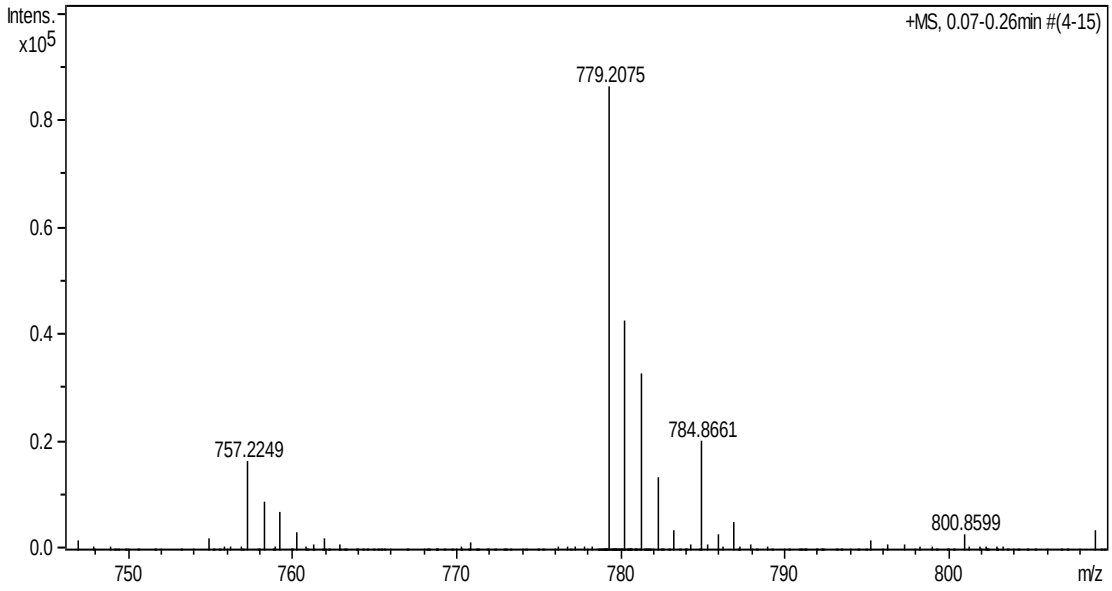
53.88
44.56
30.56
21.68
21.46
f1 (ppm)


Intens.
x10^5
+MS, 0.07-0.26min #(4-15)
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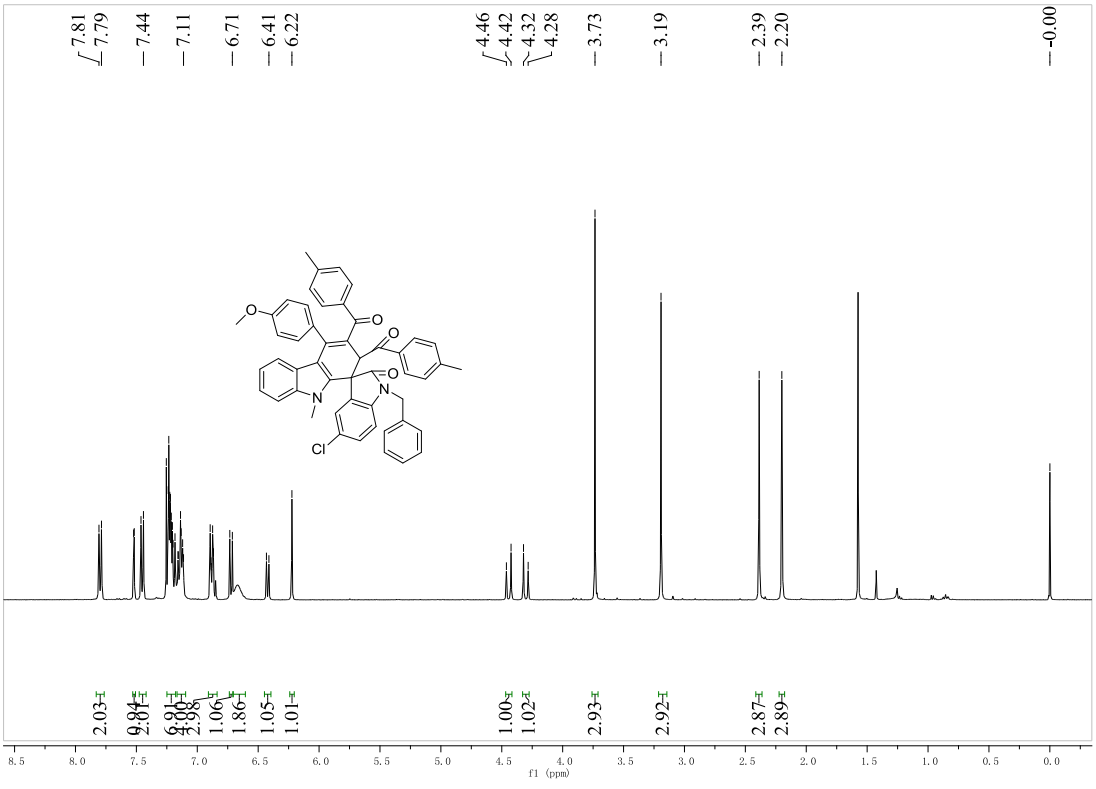
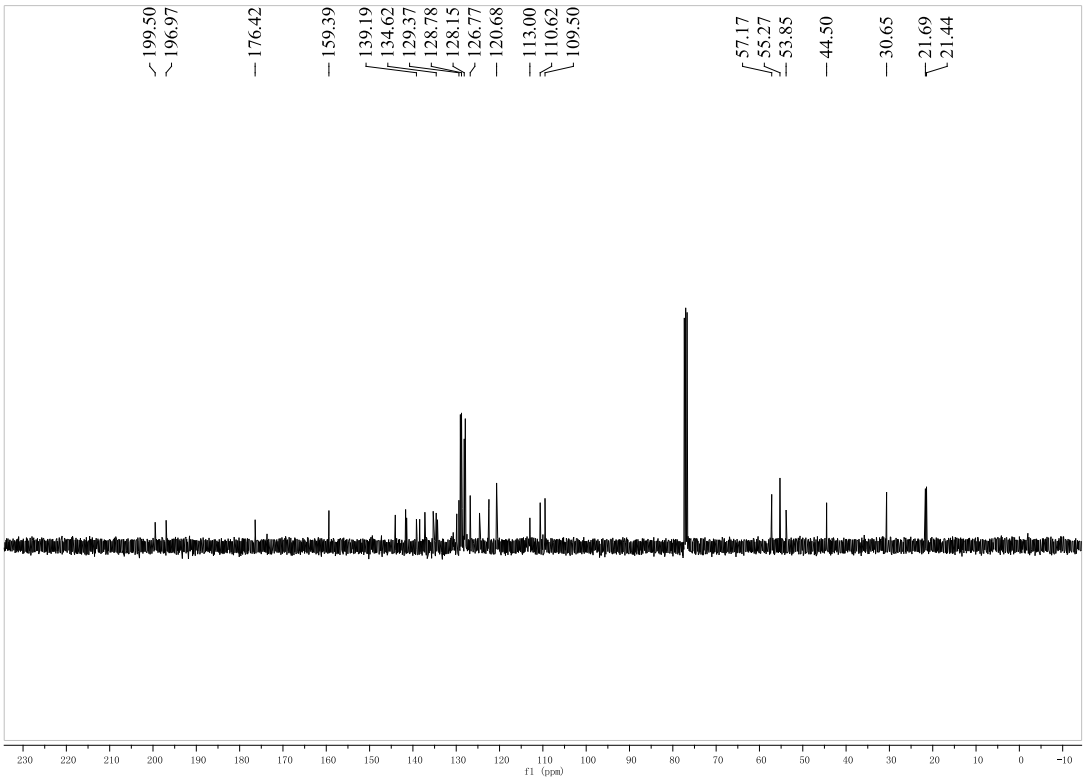
757.2249
800.8599
m/z

**(1'-Benzyl-5'-chloro-4-(4-methoxyphenyl)-9-methyl-2'-oxo-2,9-dihydrospiro[carbazole-1,3'-indoline]-2,3-diyl)bis(*p*-tolylmethanone) (3d):**

Yellow solid, 286 mg, 73%, m.p. 173-176 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.90 (d, *J* = 8.0 Hz, 2H, ArH), 7.52-7.51 (m, 1H, ArH), 7.45 (d, *J* = 8.0 Hz, 2H, ArH), 7.25-7.18 (m, 8H, ArH), 7.16-7.11 (m, 4H, ArH), 6.89-6.87 (m, 3H, ArH), 6.72 (d, *J* = 8.0 Hz, 1H, ArH), 6.67-6.64 (m, 2H, ArH), 6.42 (d, *J* = 8.0 Hz, 1H, ArH), 6.22 (s, 1H, CH), 4.44 (d, *J* = 16.0 Hz, 1H, CH), 4.30 (d, *J* = 16.0 Hz, 1H, CH), 3.74 (s, 3H, $OCH_3$), 3.19 (s, 3H, $CH_3$), 2.39 (s, 3H, $CH_3$), 2.20 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 199.5, 196.9, 176.4, 159.3, 144.0, 141.6, 141.4, 139.1, 138.4, 137.2, 135.3, 134.6, 134.3, 129.8, 129.4, 129.3, 129.2, 129.1, 129.0, 128.8, 128.7, 128.6, 128.2, 128.1, 128.0, 127.9, 126.8, 126.7, 124.6, 122.4, 120.7, 120.6, 113.0, 110.6, 109.5, 57.1, 55.2, 53.8, 44.5, 30.6, 21.7, 21.4; IR(KBr) υ: 3054, 2932, 1717, 1645, 1599, 1507, 1466, 1304, 1250, 1214, 1176, 1120, 1037, 871, 827, 750, 620, 477 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{50}H_{39}ClN_2O_4$ ($[M+Na]^+$): 785.2491, found: 785.3360.




199.50
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134.62
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128.78
128.15
126.77
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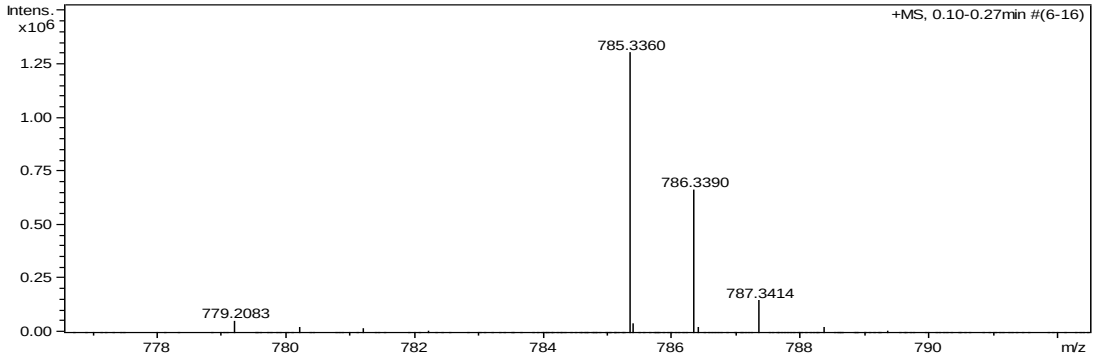
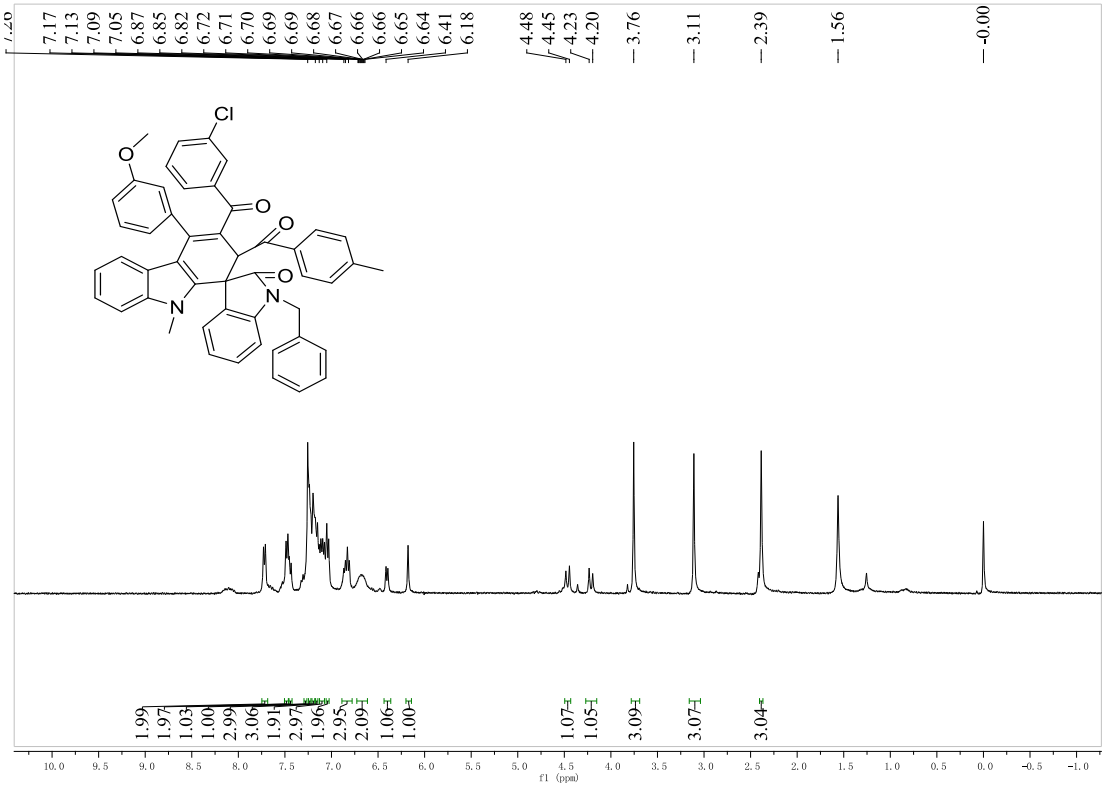
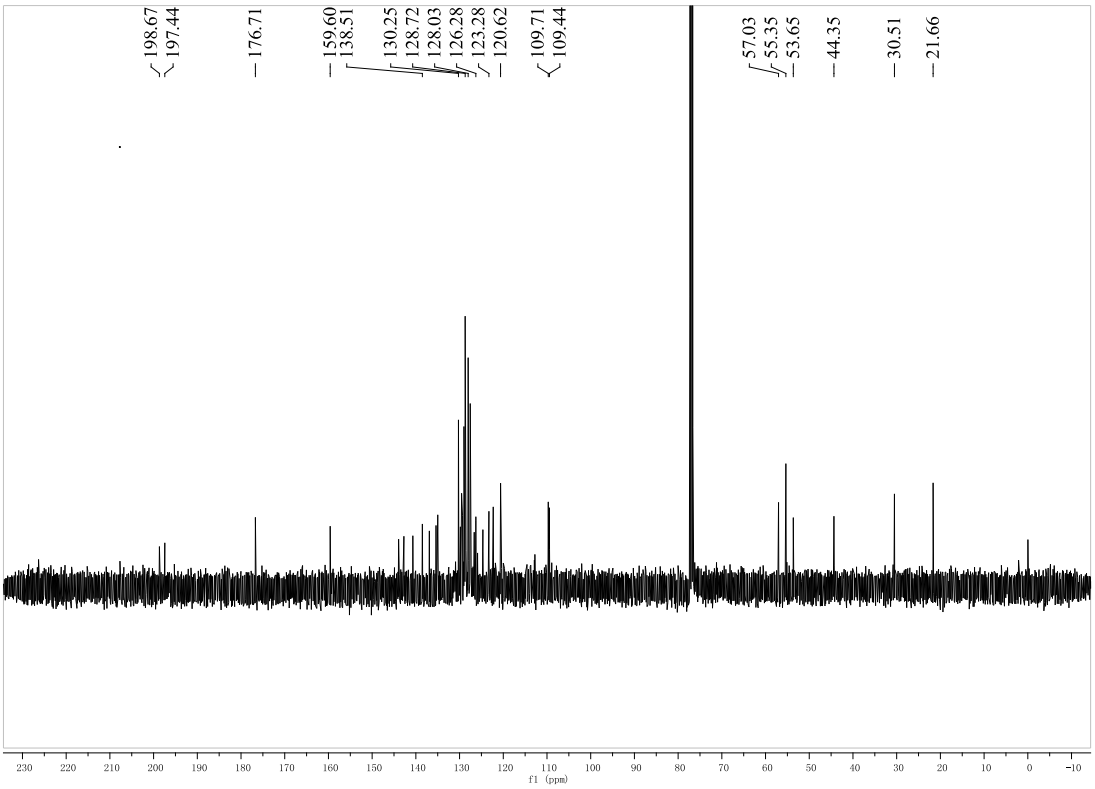
53.85
44.50
30.65
21.69
21.44
f1 (ppm)


Intens.
x10^6
+MS, 0.10-0.27min #(6-16)
785.3360
786.3390
787.3414
779.2083
m/z

**1'-Benzyl-3-(3-chlorobenzoyl)-4-(3-methoxyphenyl)-9-methyl-2-(4-methylbenzoyl)-2,9-dihydrospiro[carbazole-1,3'-indolin]-2'-one (3e):**

Yellow solid, 302 mg, 78%, m.p. 171-173 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.72 (d, *J* = 8.0 Hz, 2H, ArH), 7.47 (d, *J* = 8.0 Hz, 2H, ArH), 7.44 (d, *J* = 8.0 Hz, 1H, ArH), 7.30-7.26 (m, 1H, ArH), 7.24-7.22 (m, 3H, ArH), 7.20-7.18 (m, 3H, ArH), 7.17-7.15 (m, 2H, ArH), 7.13-7.08 (m, 3H, ArH), 7.04 (d, *J* = 8.4 Hz, 2H, ArH), 6.87-6.81 (m, 3H, ArH), 6.72-6.64 (m, 2H, ArH), 6.40 (d, *J* = 7.6 Hz, 1H, ArH), 6.18 (s, 1H, CH), 4.46 (d, *J* = 15.2 Hz, 1H, CH), 4.21 (d, *J* = 15.2 Hz, 1H, CH), 3.76 (s, 3H, $OCH_3$), 3.11 (s, 3H, $CH_3$), 2.39 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 198.6, 197.4, 176.7, 159.5, 143.9, 142.7, 140.6, 138.5, 136.9, 135.3, 135.0, 134.9, 130.2, 128.9, 128.7, 128.6, 128.0, 127.5, 126.2, 124.6, 123.2, 122.2, 120.6, 109.7, 109.4, 57.0, 55.3, 53.6, 44.3, 30.5, 21.6; IR(KBr) υ: 3047, 2983, 1766, 1612, 1571, 1483, 1474, 1362, 1255, 1238, 1167, 1134, 1052, 899, 753, 683, 655 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{49}H_{37}ClN_2O_4$ ($[M+Na]^+$): 775.2334, found: 775.2320.

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197.44
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159.60
138.51
130.25
128.72
128.03
126.28
123.28
120.62
109.71
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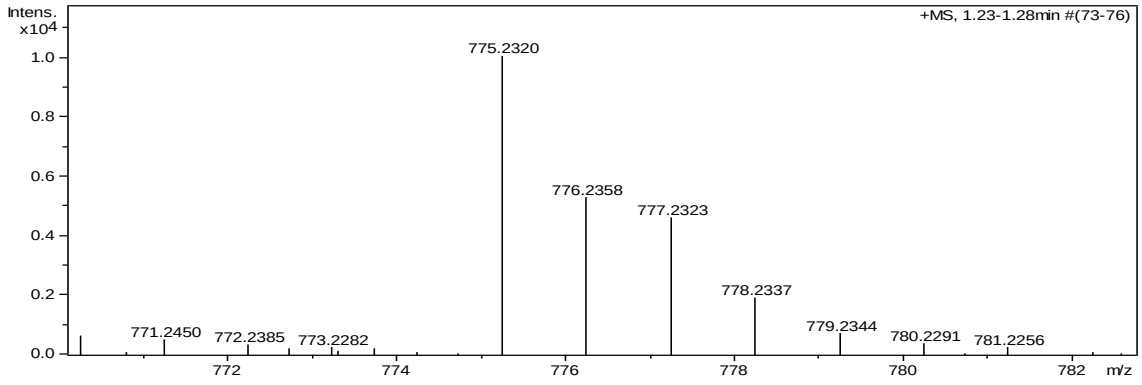
55.35
53.65
44.35
30.51
21.66
f1 (ppm)


Intens.
x10^4
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776.2358
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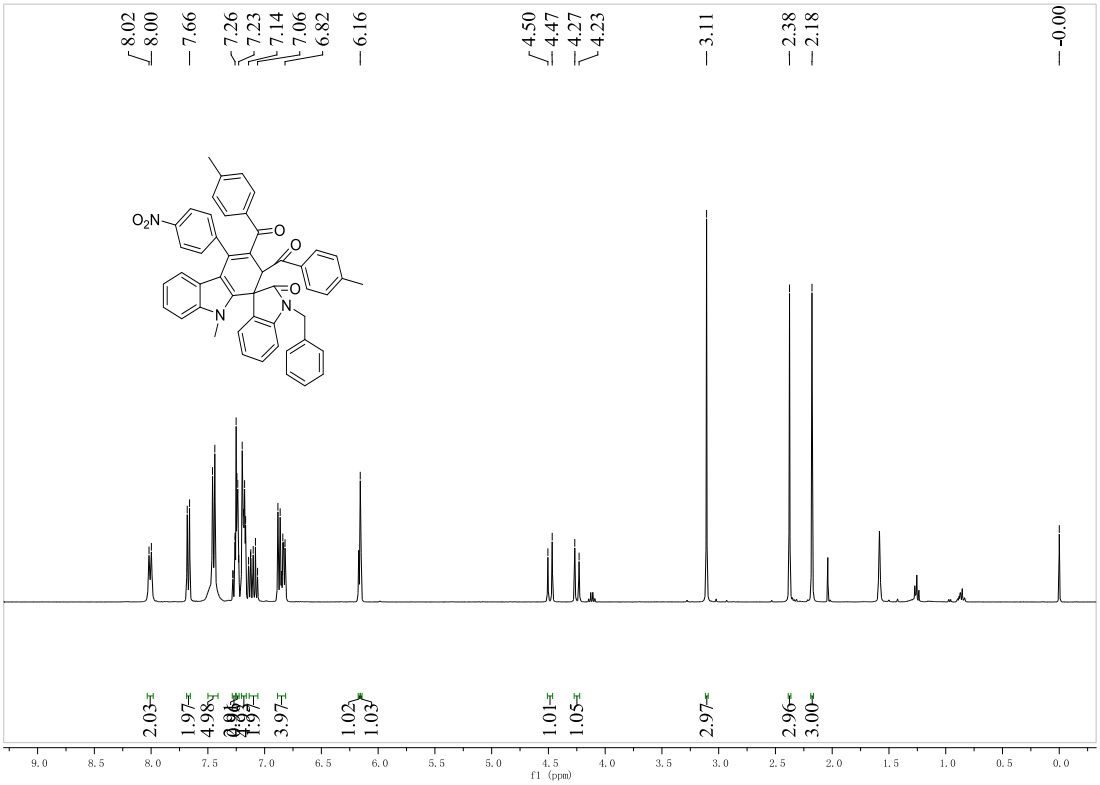
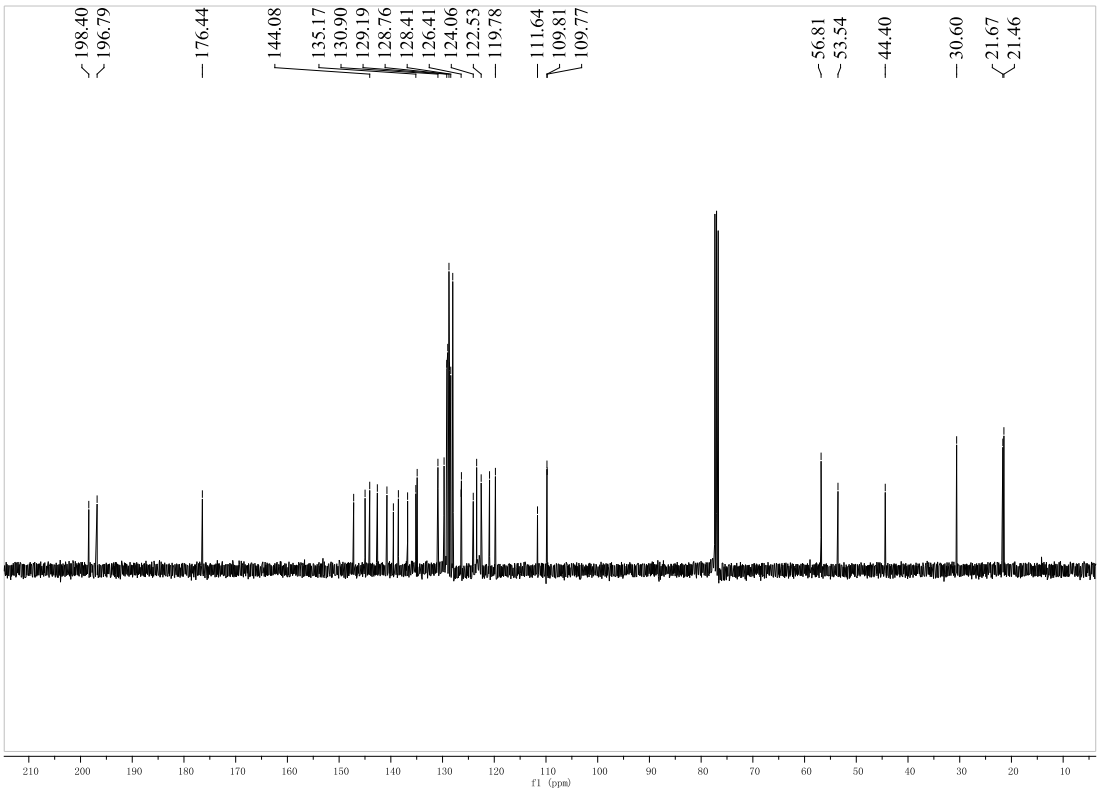
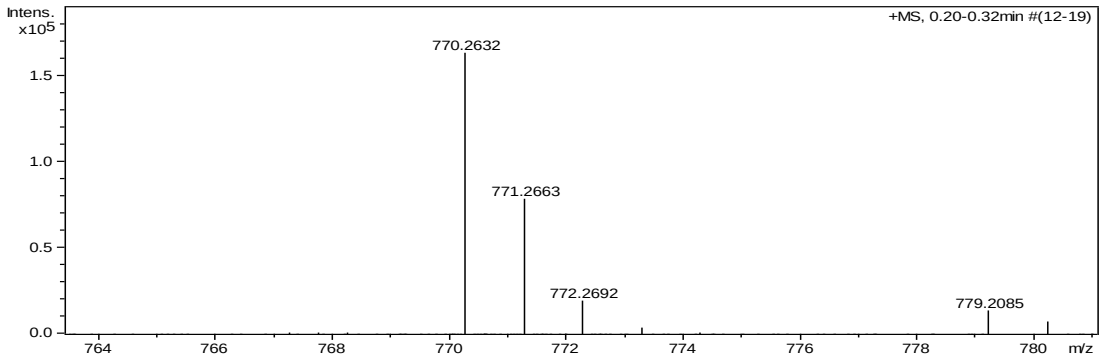
778.2337
779.2344
780.2291
781.2256
771.2450
772.2385
773.2282
m/z

**(1'-Benzyl-9-methyl-4-(4-nitrophenyl)-2'-oxo-2,9-dihydrospiro[carbazole-1,3'-indoline]-2,3-diyl)bis(*p*-tolylmethanone) (3f):**

Yellow solid, 285 mg, 74%, m.p. 168-170 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.81 (d, *J* = 8.0 Hz, 2H, ArH), 7.67 (d, *J* = 8.0 Hz, 2H, ArH), 7.46-7.44 (m, 5H, ArH), 7.28-7.25 (m, 1H, ArH), 7.25-7.23 (m, 3H, ArH), 7.20-7.17 (m, 5H, ArH), 7.14-7.06 (m, 2H, ArH), 6.88-6.81 (m, 4H, ArH), 6.17-6.16 (m, 1H, ArH), 6.16-6.15 (m, 1H, CH), 4.49 (d, *J* = 15.2 Hz, 1H, CH), 4.25 (d, *J* = 15.2 Hz, 1H, CH), 3.11 (s, 3H, $CH_3$), 2.38 (s, 3H, $CH_3$), 2.18 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 198.4, 196.7, 176.4, 147.1, 144.9, 144.0, 142.6, 140.7, 139.5, 138.5, 136.7, 135.1, 134.9, 130.8, 129.7, 128.6, 128.4, 128.0, 126.4, 126.3, 124.0, 123.4, 122.5, 120.9, 119.7, 111.6, 109.8, 109.7, 56.8, 53.5, 44.4, 30.5, 21.6, 21.4; IR(KBr) υ: 3060, 2912, 1715, 1600, 1516, 1467, 1409, 1335, 1268, 1224, 1182, 1113, 1009, 824, 743, 698, 626, 486 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{49}H_{37}N_3O_5$ ([M+Na]$^+$): 770.2625, found: 770.2632.

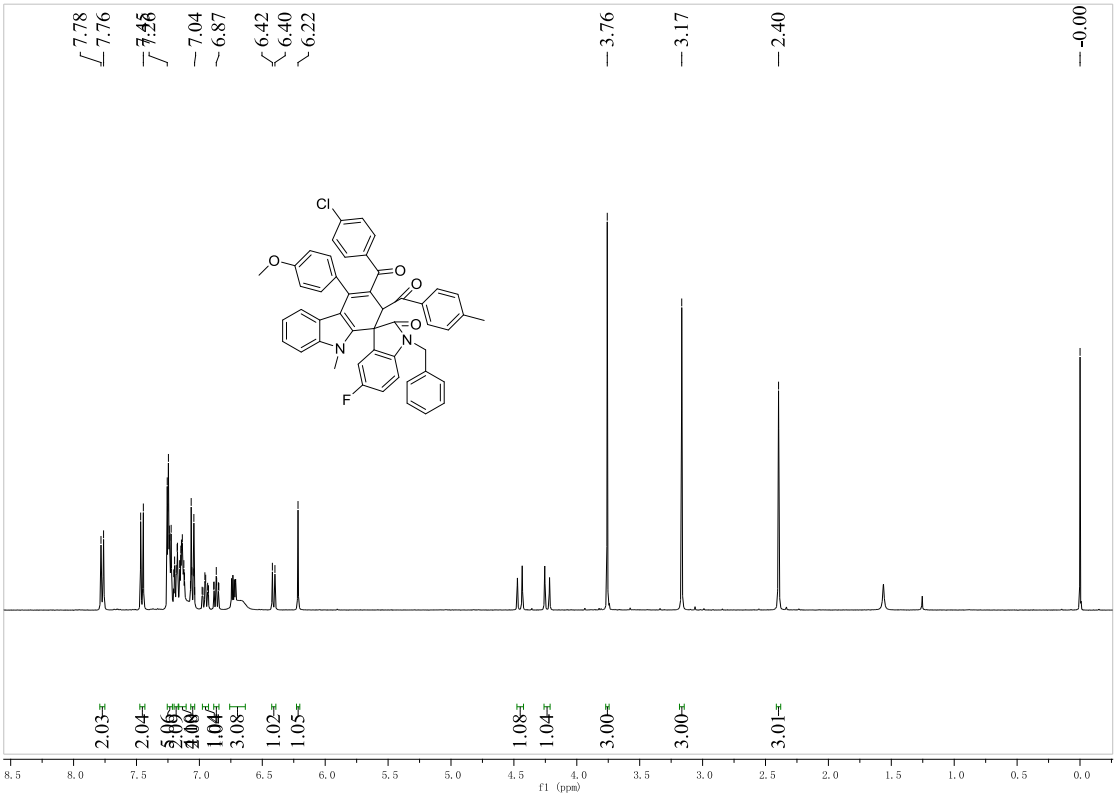


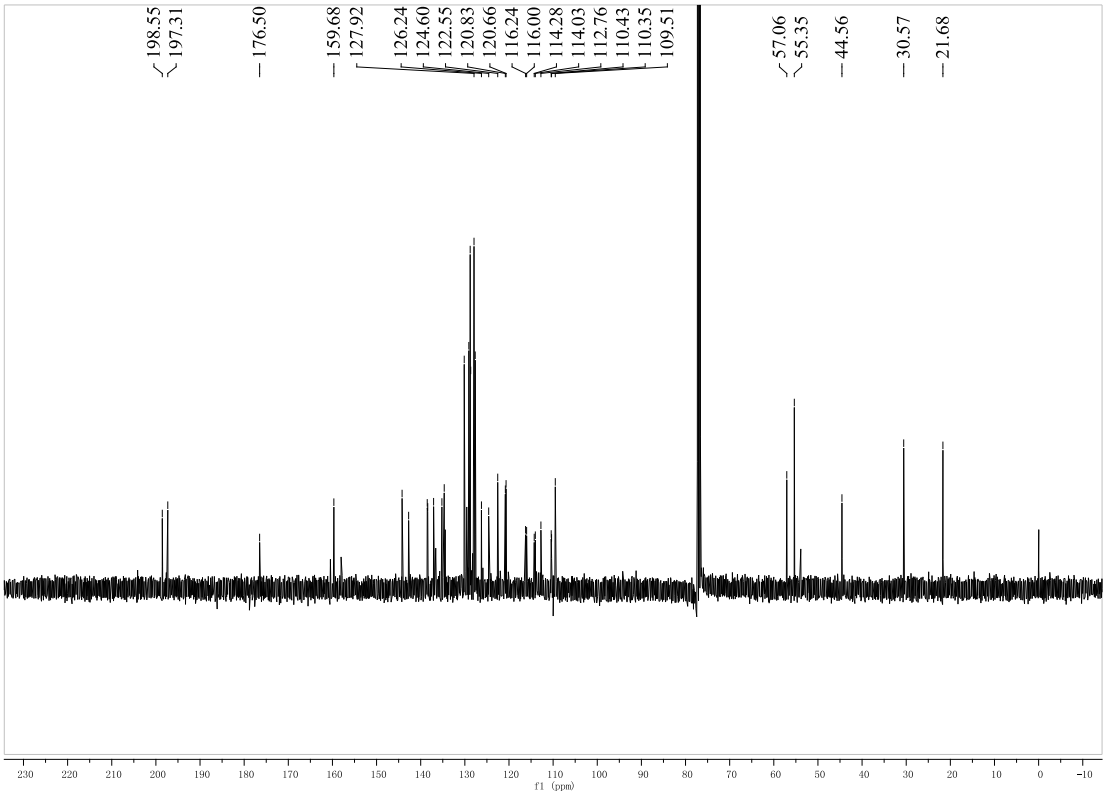

198.40
196.79
176.44
144.08
135.17
130.90
129.19
128.76
128.41
126.41
124.06
122.53
119.78
111.64
109.81
109.77
56.81
53.54
44.40
30.60
21.67
21.46
f1 (ppm)

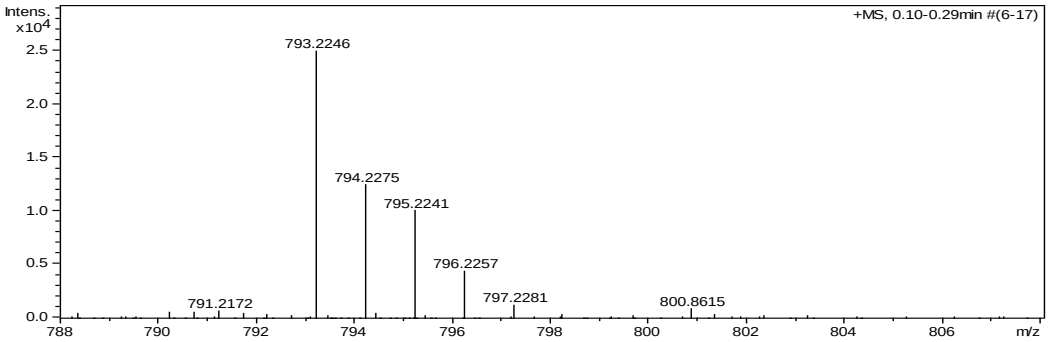

Intens.
x10^5
+MS, 0.20-0.32min #(12-19)
770.2632
771.2663
772.2692
779.2085
m/z

**1'-Benzyl-3-(4-chlorobenzoyl)-5'-fluoro-4-(4-methoxyphenyl)-9-methyl-2-(4-methylbenzoyl)-2,9-dihydrospiro[carbazole-1,3'-indolin]-2'-one (3g):**

Yellow solid, 282 mg, 71%, m.p. 175-178 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.77 (d, *J* = 8.0 Hz, 2H, ArH), 7.46 (d, *J* = 8.0 Hz, 2H, ArH), 7.26-7.20 (m, 6H, ArH), 7.20-7.18 (m, 2H, ArH), 7.16-7.12 (m, 4H, ArH), 7.07-7.04 (m, 2H, ArH), 6.98-6.93 (m, 1H, ArH), 6.89-6.85 (m, 1H, ArH), 6.74-6.66 (m, 3H, ArH), 6.41 (d, *J* = 8.0 Hz, 1H, ArH), 6.22 (s, 1H, CH), 4.45 (d, *J* = 15.2 Hz, 1H, CH), 4.24 (d, *J* = 15.2 Hz, 1H, CH), 3.76 (s, 3H, $OCH_3$), 3.17 (s, 3H, $CH_3$), 2.40 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 198.5, 197.3, 176.4, 159.6, 144.2, 142.7, 138.5, 138.4, 137.0, 135.1, 134.6, 130.1, 129.0, 128.7, 128.6, 128.0, 127.9, 127.6, 126.2, 124.5, 122.5, 120.8, 120.6, 116.2, 116.0, 114.2, 114.0, 112.7, 110.4, 110.3, 109.5, 57.0, 55.3, 44.5, 30.5, 21.6; IR(KBr) υ: 3061, 2902, 1715, 1599, 1479, 1405, 1320, 1248, 1179, 1122, 1017, 952, 806, 745, 698, 609, 561 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{49}H_{36}ClFN_2O_4$ ([M+Na]$^{+}$): 793.2240, found: 793.2246.

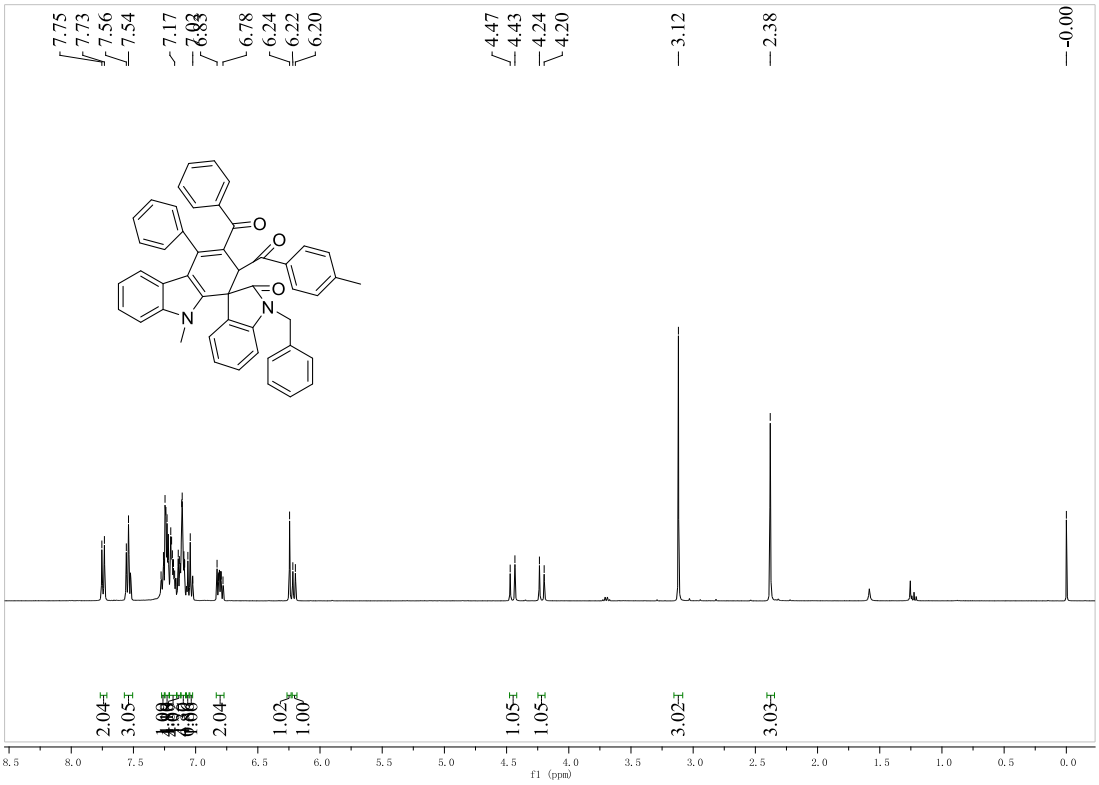


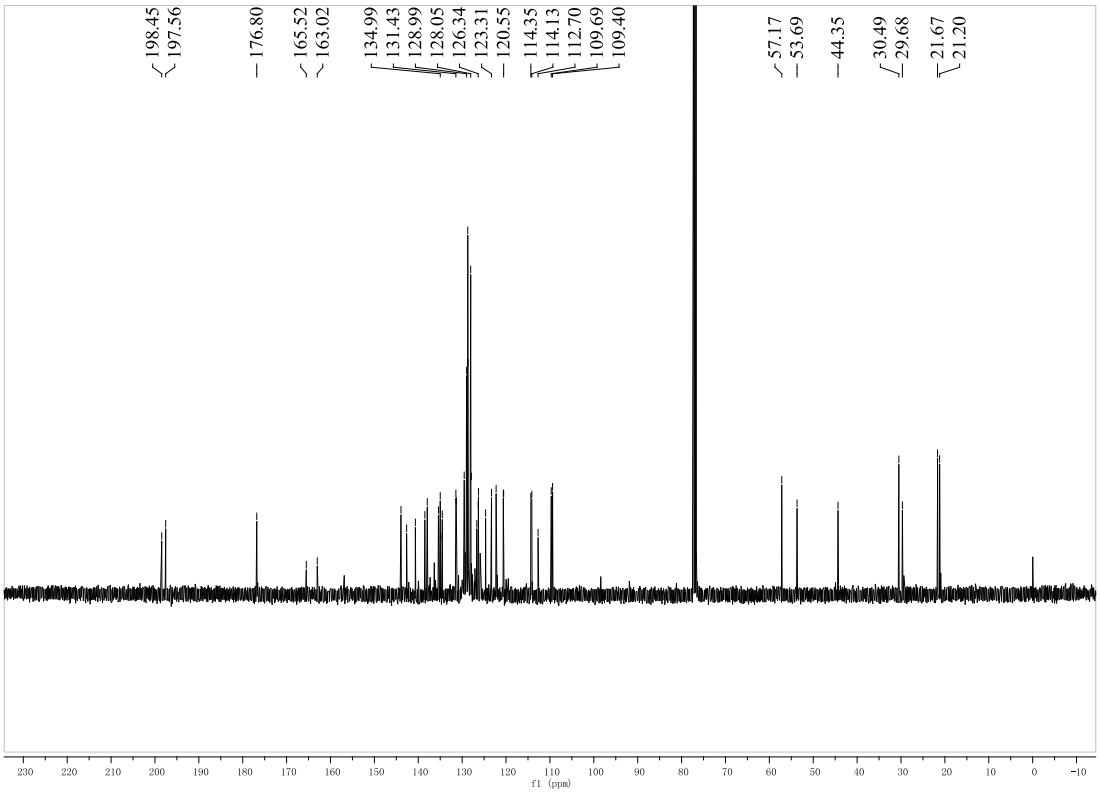

198.55
197.31
176.50
159.68
127.92
126.24
124.60
122.55
120.83
120.66
116.24
116.00
114.28
114.03
112.76
110.43
110.35
109.51
57.06
55.35
44.56
30.57
21.68
f1 (ppm)


Intens.
x10^4
+MS, 0.10-0.29min #(6-17)
793.2246
794.2275
795.2241
796.2257
797.2281
791.2172
800.8615
m/z

**3-Benzoyl-1'-benzyl-9-methyl-2-(4-methylbenzoyl)-4-phenyl-2,9-dihydrospiro[carbazole-1,3'-indolin]-2'-one (3h):**

Yellow solid, 292 mg, 82%, m.p. 160-162 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.74 (d, *J* = 8.0 Hz, 2H, ArH), 7.56-7.52 (m, 3H, ArH), 7.28-7.26 (m, 2H, ArH), 7.24-7.22 (m, 4H, ArH), 7.20-7.16 (m, 4H, ArH), 7.15-7.07 (m, 7H, ArH), 7.06-7.03 (m, 2H, ArH), 6.83-6.78 (m, 2H, ArH), 6.25 (s, 1H, CH), 6.21 (d, *J* = 8.0 Hz, 1H, ArH), 4.45 (d, *J* = 15.2 Hz, 1H, CH), 4.22 (d, *J* = 15.2 Hz, 1H, CH), 3.12 (s, 3H, $CH_3$), 2.38 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 198.4, 197.5, 176.8, 165.5, 163.0, 143.9, 142.6, 140.6, 138.4, 137.9, 135.4, 134.5, 131.4, 129.5, 128.9, 128.0, 127.9, 126.6, 126.3, 124.6, 123.3, 122.2, 120.5, 114.3, 114.1, 112.6, 109.6, 109.3, 57.1, 53.6, 44.3, 30.4, 29.6, 21.6, 21.2; IR(KBr) υ: 3037, 2926, 1894, 1713, 1679, 1612, 1507, 1482, 1406, 1321, 1328, 1229, 1119, 1012, 912, 848, 809, 741, 693, 637, 559 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{48}H_{36}N_2O_3$ ([M+Na]$^+$): 711.2618, found: 711.2641.




198.45
197.56
176.80
165.52
163.02
134.99
131.43
128.99
128.05
126.34
123.31
120.55
114.35
114.13
112.70
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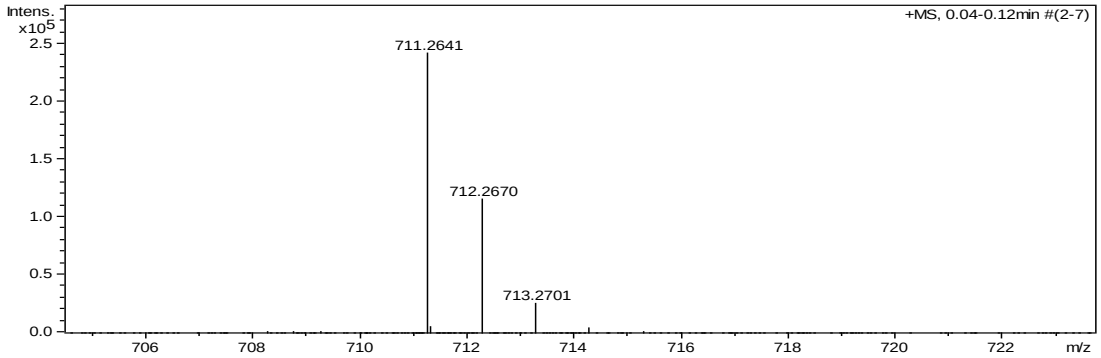
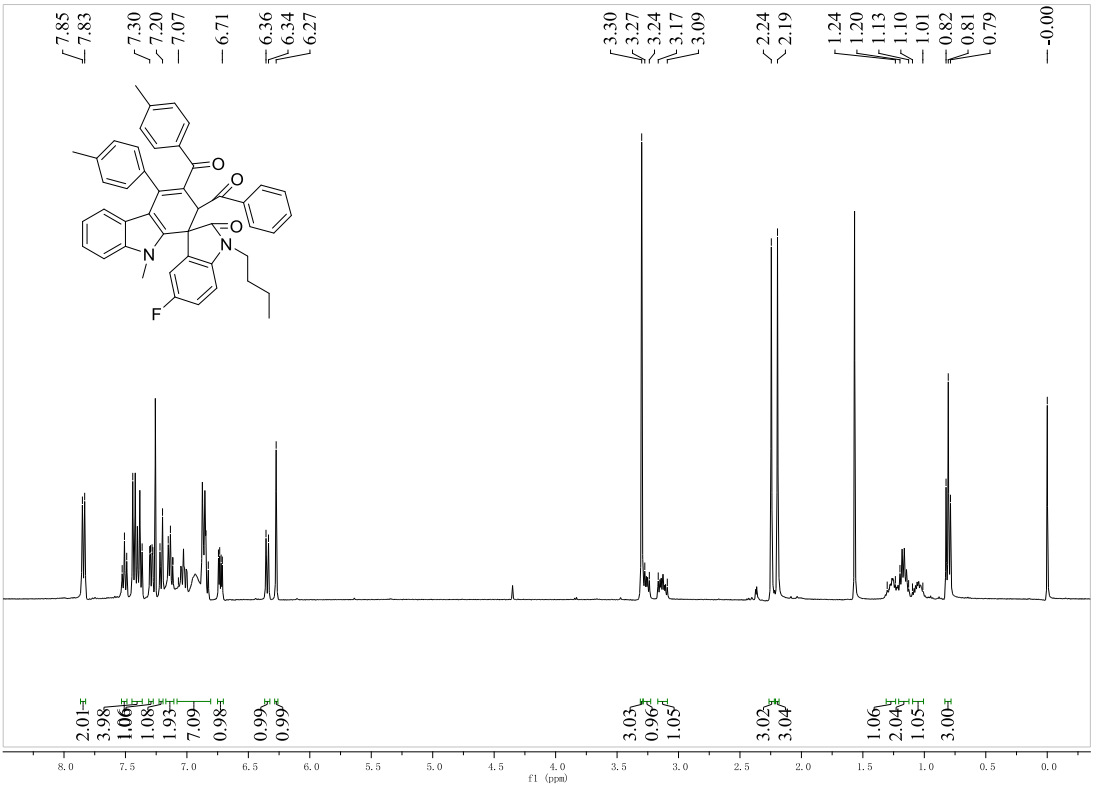
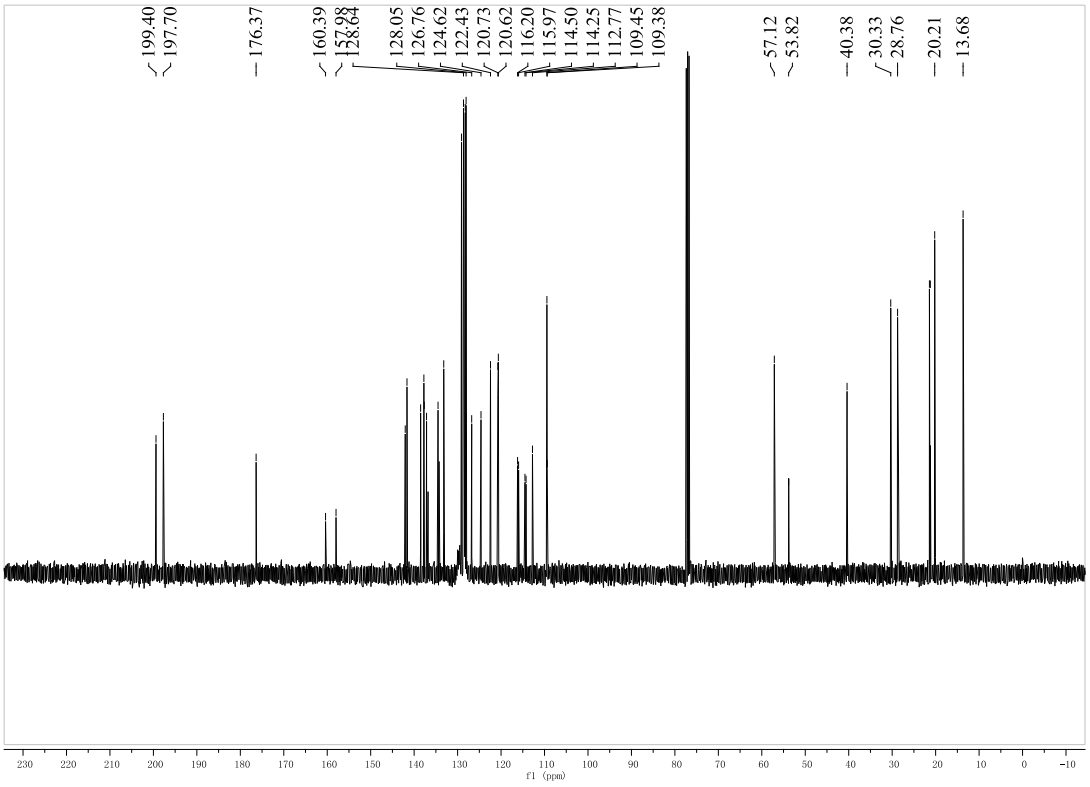
109.40
57.17
53.69
44.35
30.49
29.68
21.67
21.20
f1 (ppm)

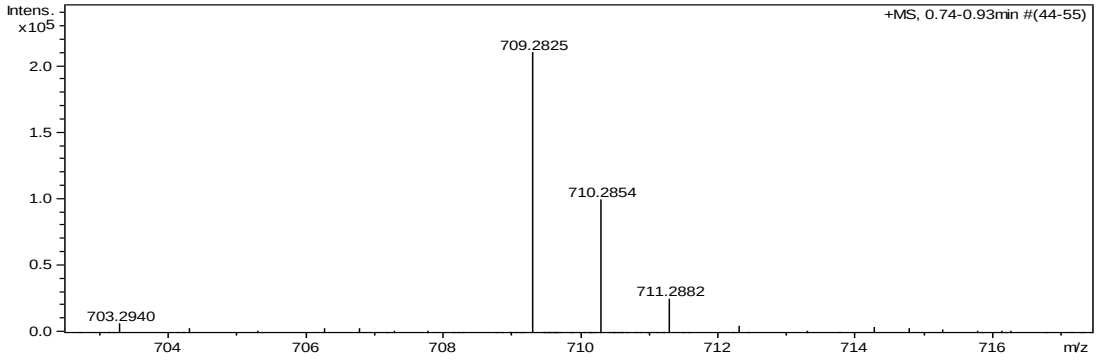

Intens.
x10^5
+MS, 0.04-0.12min #(2-7)
711.2641
712.2670
713.2701
m/z

**2-Benzoyl-1'-butyl-5'-fluoro-9-methyl-3-(4-methylbenzoyl)-4-(*p*-tolyl)-2,9-dihydrospiro[carbazole-1,3'-indolin]-2'-one (3i):**

Yellow solid, 220 mg, 62%, m.p. 177-180 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.84 (d, *J* = 7.2 Hz, 2H, ArH), 7.51 (t, *J* = 7.6 Hz, 1H, ArH), 7.44-7.36 (m, 4H, ArH), 7.29 (d, d, $J_1$ = 7.6 Hz, $J_2$ = 2.8 Hz, 1H, ArH), 7.21 (d, *J* = 8.4 Hz, 1H, ArH), 7.15-7.11 (m, 2H, ArH), 7.06-6.82 (m, 7H, ArH), 6.73 (d, d, $J_1$ = 8.4 Hz, $J_2$ = 4.0 Hz, 1H, ArH), 6.35 (d, *J* = 8.4 Hz, 1H, ArH), 6.27 (s, 1H, CH), 3.30 (s, 3H, $OCH_3$), 3.28-3.24 (m, 1H, CH), 3.17-3.09 (m, 1H, CH), 2.25 (s, 3H, $CH_3$), 2.20 (s, 3H, $CH_3$), 1.30-1.24 (m, 1H, CH), 1.20-1.13 (m, 2H, $CH_2$), 1.10-1.01 (m, 1H, CH), 0.81 (d, *J* = 7.2 Hz, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 199.40, 197.70, 176.36, 160.38, 157.97, 142.05, 141.65, 138.50, 137.76, 137.67, 137.14, 134.50, 133.19, 129.09, 128.64, 126.75, 124.61, 122.43, 120.73, 115.96, 114.50, 112.77, 109.45, 57.12, 53.82, 40.38, 30.33, 28.75, 21.43, 21.21, 20.21, 13.68; IR (KBr) υ: 3045, 2924, 1895, 1717, 1597, 1508, 1466, 1408, 1318, 1267, 1221, 1185, 1151, 1009, 906, 832, 796, 744, 695, 556 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{47}H_{41}FN_2O_3$ ($[M+Na]^+$): 709.2837, found: 709.2825.

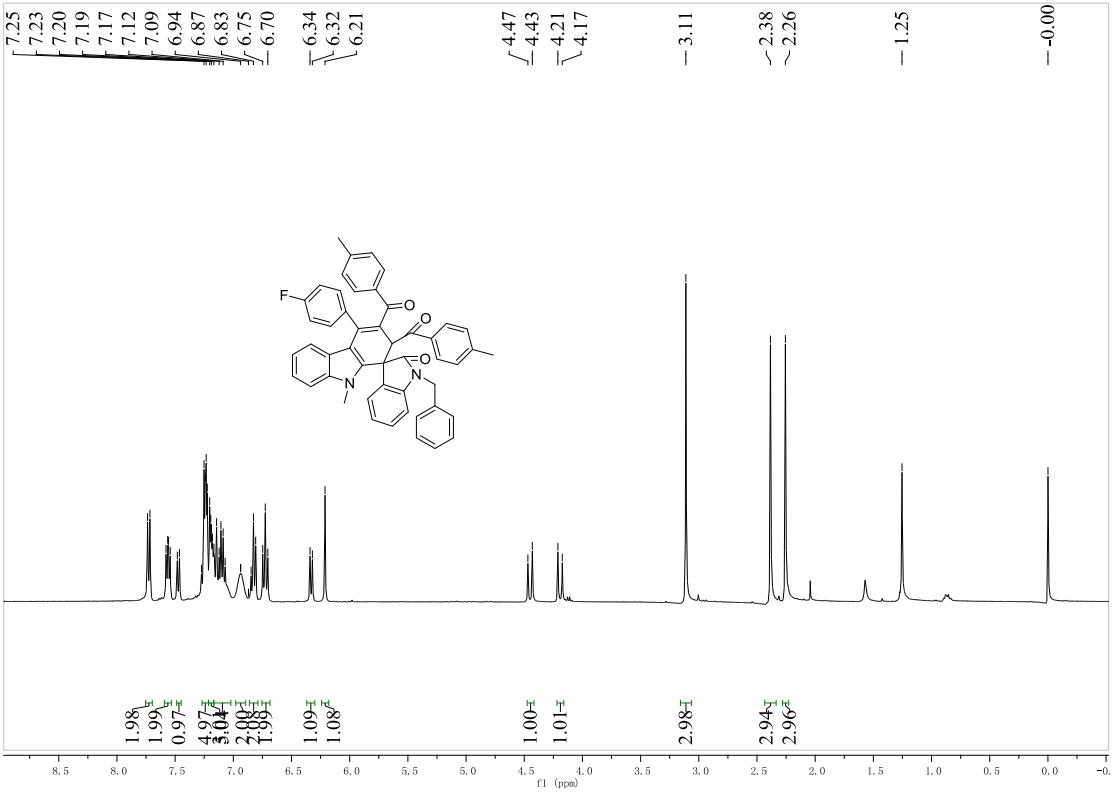


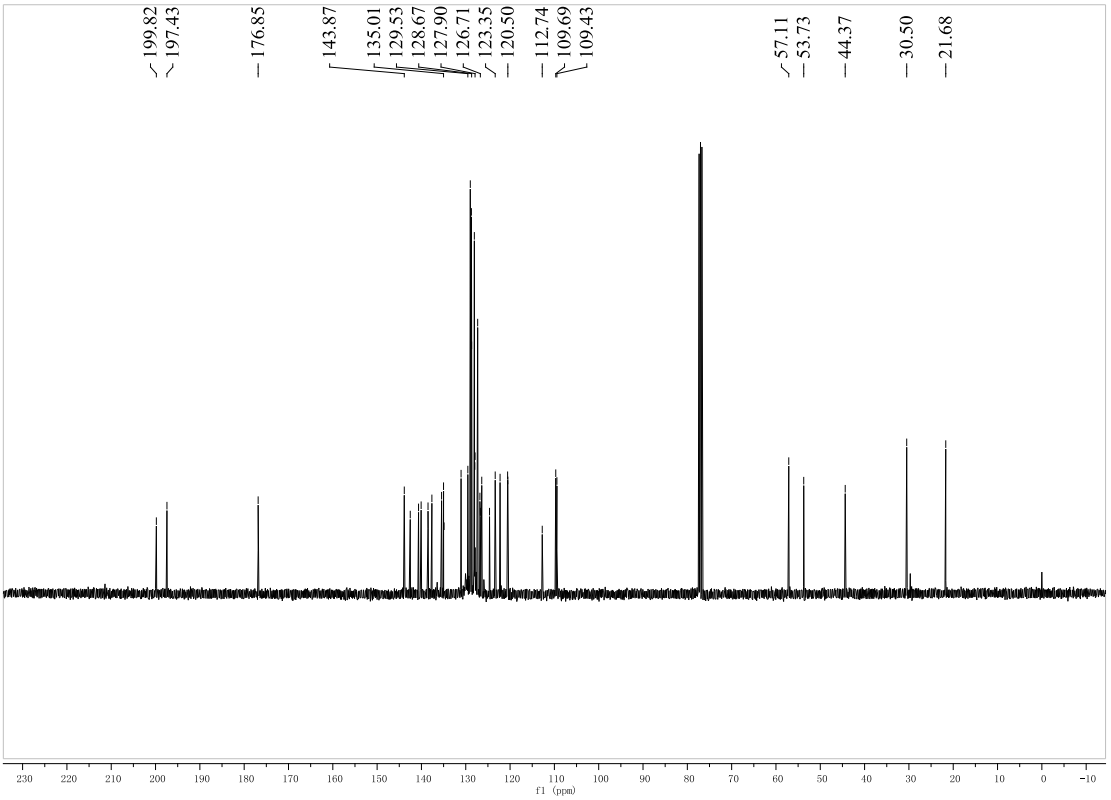

199.40
197.70
176.37
160.39
157.98
128.84
128.05
126.76
124.62
122.43
120.73
120.62
116.20
115.97
114.50
114.25
112.77
109.45
109.38
57.12
53.82
40.38
30.33
28.76
20.21
13.68
f1 (ppm)

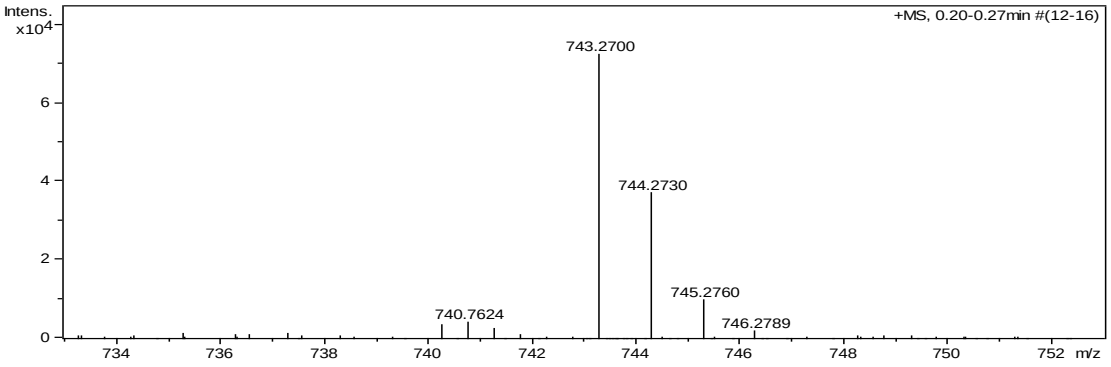

Intens.
x10^5
+MS, 0.74-0.93min #(44-55)
709.2825
710.2854
711.2882
703.2940
m/z

**(1'-Benzyl-4-(4-fluorophenyl)-9-methyl-2'-oxo-2,9-dihydrospiro[carbazole-1,3'-indoline]-2,3-diyl)bis(*p*-tolylmethanone) (3j):**

Yellow solid, 286 mg, 77%, m.p. 171-173 °C;$^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.73 (d, *J* = 8.0 Hz, 2H, ArH), 7.58-7.54 (m, 2H, ArH), 7.47 (d, *J* = 7.2 Hz, 1H, ArH), 7.25-7.22 (m, 5H, ArH), 7.20-7.18 (m, 3H, ArH), 7.14-7.07 (m, 5H, ArH), 6.94-6.87 (m, 2H, ArH), 6.85-6.81 (m, 2H, ArH), 6.75-6.71 (m, 2H, ArH), 6.33 (d, *J* = 8.0 Hz, 1H, ArH), 6.21 (s, 1H, CH), 4.45 (d, *J* = 15.2 Hz, 1H, CH), 4.19 (d, *J* = 15.2 Hz, 1H, CH), 3.11 (s, 3H, $CH_3$), 2.39 (s, 3H, $CH_3$), 2.26 (s, 3H, $CH_3$);$^{13}$C NMR (100 MHz, $CDCl_3$) δ: 199.8, 197.4, 176.8, 143.8, 142.5, 140.6, 140.0, 138.5, 137.6, 135.4, 135.0, 134.8, 131.0, 129.5, 128.9, 128.7, 128.6, 128.0, 127.9, 127.3, 126.8, 126.7, 126.3, 124.6, 123.3, 122.2, 120.5, 112.7, 109.6, 109.4, 57.1, 53.7, 44.3, 30.4, 21.6; IR(KBr) υ: 3045, 2924, 1895, 1717, 1597, 1508，1466, 1408, 1318, 1267, 1221，1185, 1151, 1009, 906, 832, 796, 744，695，556 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{49}H_{37}FN_2O_3$ ([M+Na]$^{+}$): 743.2680, found: 743.2700.

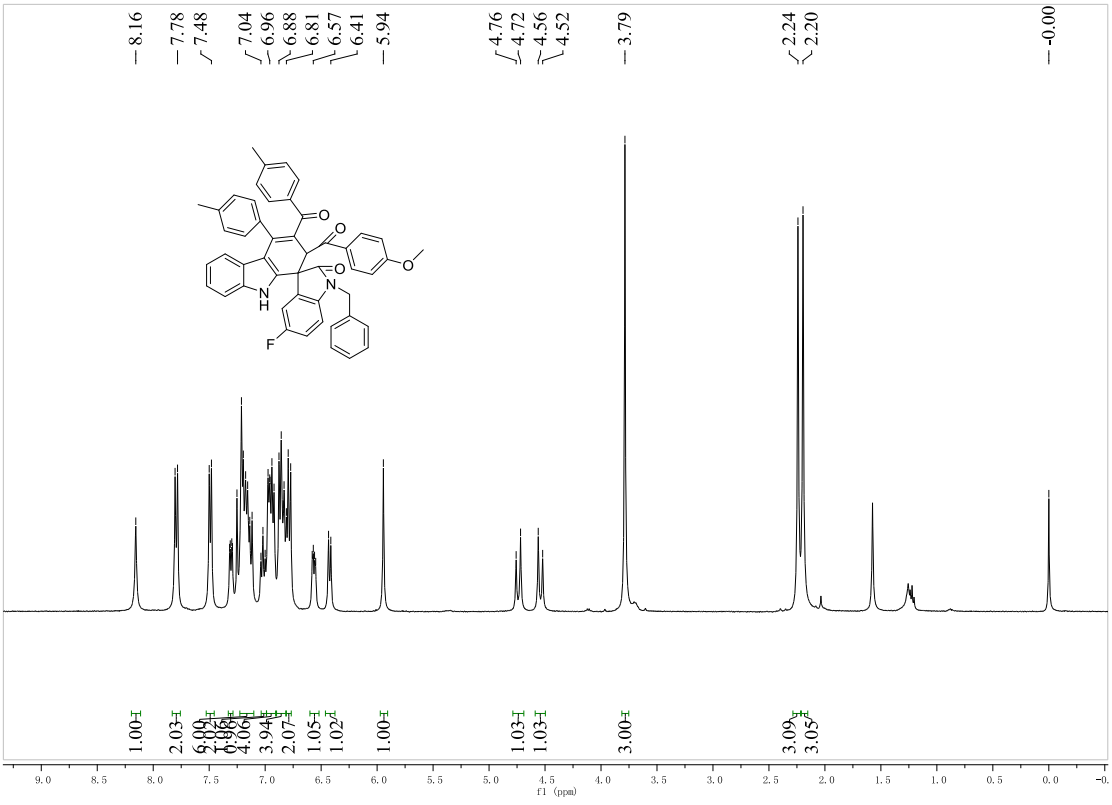


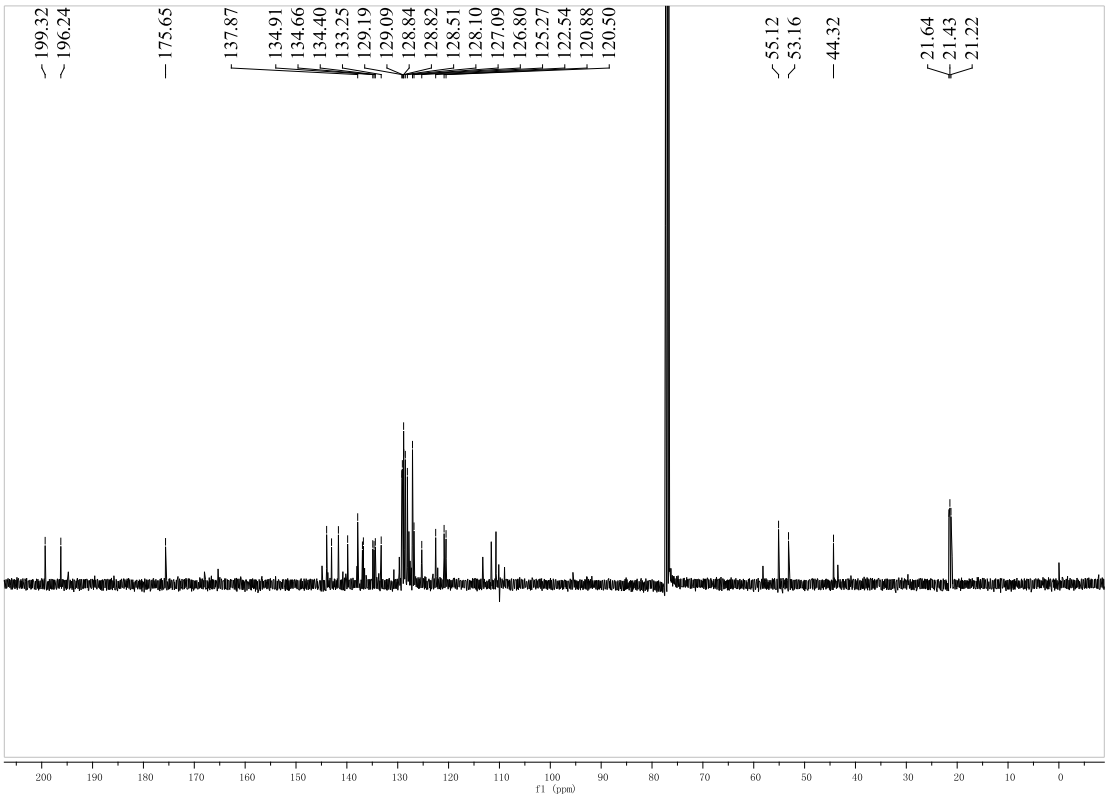

199.82
197.43
176.85
143.87
135.01
129.53
128.67
127.90
126.71
123.35
120.50
112.74
109.69
109.43
57.11
53.73
44.37
30.50
21.68
f1 (ppm)


Intens.
x10^4
+MS, 0.20-0.27min #(12-16)
743.2700
744.2730
745.2760
746.2789
740.7624
m/z

**1'-Benzyl-5'-fluoro-2-(4-methoxybenzoyl)-3-(4-methylbenzoyl)-4-(*p*-tolyl)-2,9-dihydrospiro[carbazole-1,3'-indolin]-2'-one (3k):**

Yellow solid, 315 mg, 83%, m.p. 173-176 °C; $^1$H NMR (400 MHz, $CDCl_3$) δ: 8.16 (s, 1H, NH), 7.79 (d, *J* = 8.0 Hz, 2H, ArH), 7.49 (d, *J* = 8.0 Hz, 2H, ArH), 7.32-7.29 (m, 1H, ArH), 7.21-7.12 (m, 6H, ArH), 7.04-7.01 (m, 1H, ArH), 6.97-6.93 (m, 4H, ArH), 6.88-6.83 (m, 4H, ArH), 6.81-6.77 (m, 2H, ArH), 6.58-6.55 (m, 1H, ArH), 6.42 (d, *J* = 8.0 Hz, 1H, ArH), 5.94 (s, 1H, CH), 4.74 (d, *J* = 15.6 Hz, 1H, CH), 4.54 (d, *J* = 15.6 Hz, 1H, CH), 3.79 (s, 3H, $OCH_3$), 2.24 (s, 3H, $CH_3$), 2.20 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 199.3, 196.2, 175.6, 143.9, 143.0, 141.6, 139.8, 137.8, 134.9, 134.6, 134.4, 133.2, 129.6, 128.8, 128.7, 128.5, 128.4, 128.1, 127.9, 127.8, 126.8, 125.3, 125.2, 122.5, 120.8, 120.4, 111.6, 110.6, 110.1, 109.9, 58.1, 55.1, 53.1, 44.3, 43.4, 21.4; IR (KBr) υ: 3037, 2926, 1903, 1699, 1601, 1528, 1458, 1331, 1265, 1184, 1120, 1014, 947, 809, 740, 699, 587 cm$^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{49}H_{37}FN_2O_4$ ([M+Na]$^+$): 759.2630, found: 759.2658.




199.32
196.24
175.65
137.87
134.91
134.66
134.40
133.25
129.19
129.09
128.84
128.82
128.51
128.10
127.09
126.80
125.27
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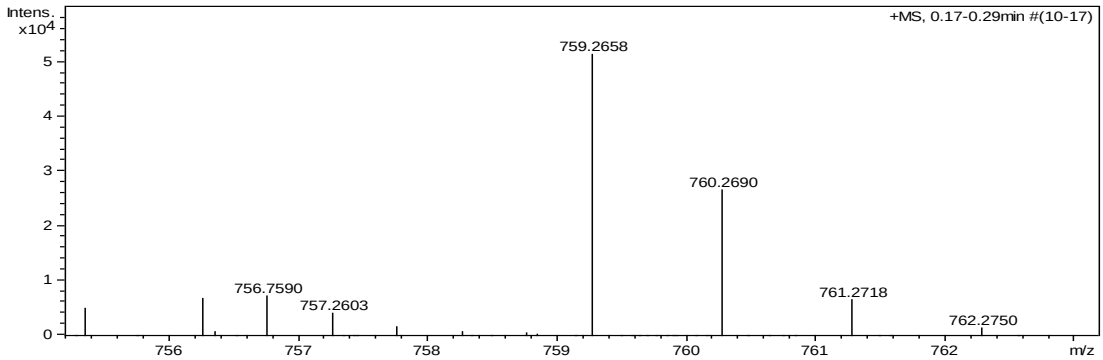
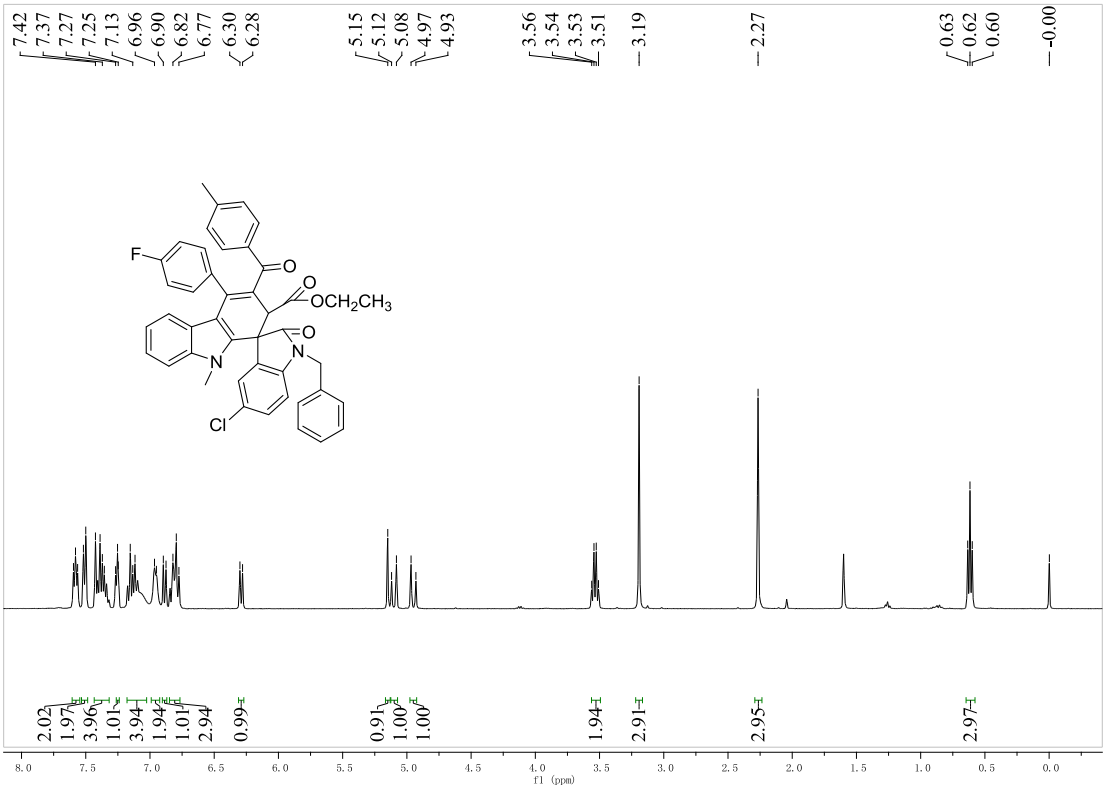
120.88
120.50
55.12
53.16
44.32
21.64
21.43
21.22
f1 (ppm)

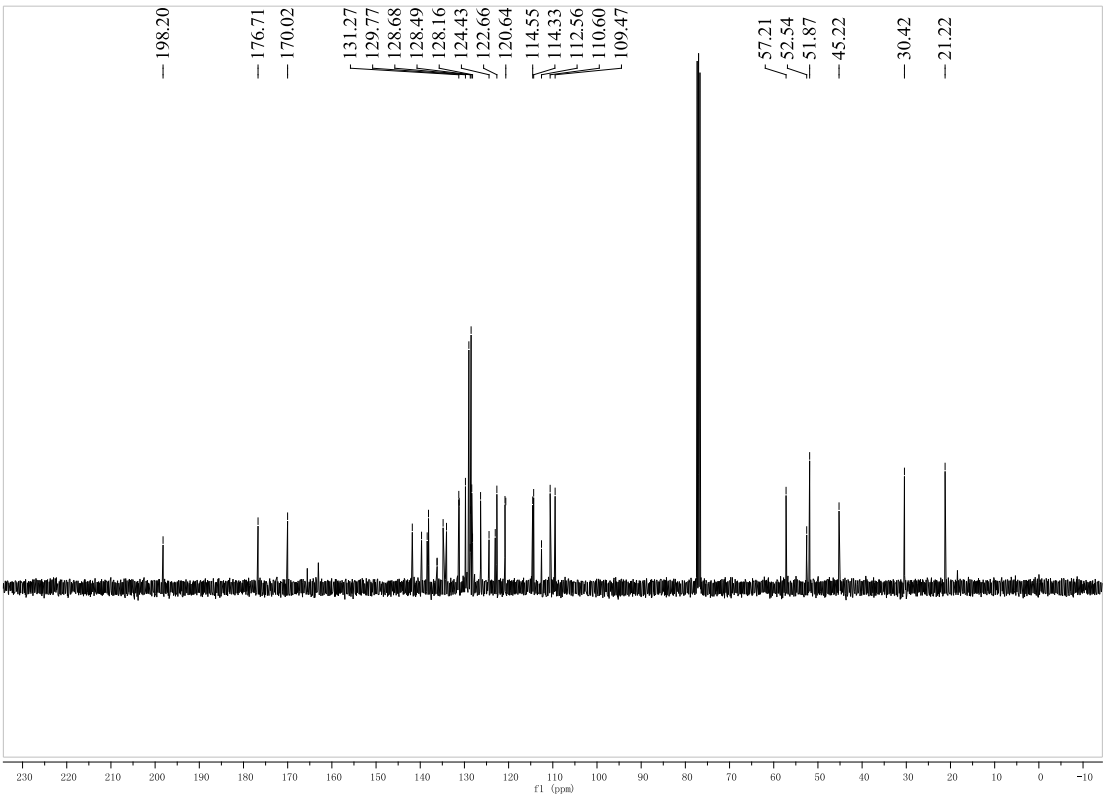

Intens.
x10^4
+MS, 0.17-0.29min #(10-17)
759.2658
760.2690
756.7590
757.2603
761.2718
762.2750
m/z

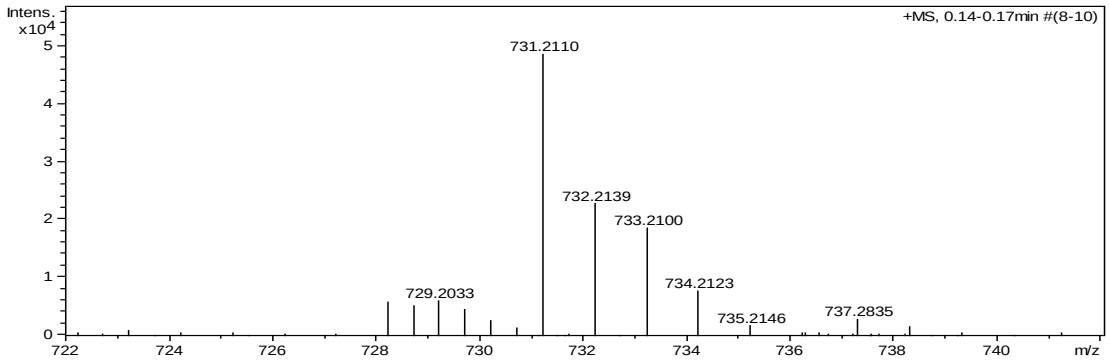
**Ethyl**

**1'-benzyl-5'-chloro-4-(4-fluorophenyl)-9-methyl-3-(4-methylbenzoyl)-2'-oxo-2,9-dihydrospiro[carbazole-1,3'-indoline]-2-carboxylate (3l):**

white solid, 292 mg, 80%, m.p. 181-184 °C; $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.60-7.57 (m, 2H, ArH), 7.51 (d, *J* = 8.0 Hz, 2H, ArH), 7.43-7.25 (m, 4H, ArH), 7.25-7.24 (m, 1H, ArH), 7.18-7.10 (m, 4H, ArH), 6.97-6.95 (m, 2H, ArH), 6.89 (d, *J* = 8.0 Hz, 1H, ArH), 6.85-6.78 (m, 3H, ArH), 6.29 (d, *J* = 8.0 Hz, 1H, ArH), 5.15 (s, 1H, CH), 5.10 (d, *J* = 15.2 Hz, 1H, CH), 4.95 (d, *J* = 15.2 Hz, 1H, CH), 3.54 (q, *J* = 7.2 Hz, 2H, $CH_2$), 3.19 (s, 3H, $CH_3$), 2.27 (s, 3H, $CH_3$), 0.62 (d, *J* = 7.2 Hz, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 198.2, 176.7, 170.0, 141.7, 139.7, 138.4, 136.2, 134.8, 131.2, 131.1, 129.7, 128.9, 128.6, 128.1, 126.3, 124.4, 123.0, 122.6, 120.8, 114.5, 112.5, 110.6, 109.4, 57.2, 52.5, 51.8, 45.2, 30.4, 21.2; IR(KBr) υ: 3056, 2982, 2935, 1897, 1724, 1637, 1597, 1510, 1472, 1416, 1323, 1270, 1227, 1184, 1026, 946, 809, 746, 698, 581 cm$^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{44}H_{34}ClFN_2O_4$ ([M+Na]$^+$): 731.2083, found:731.2110.



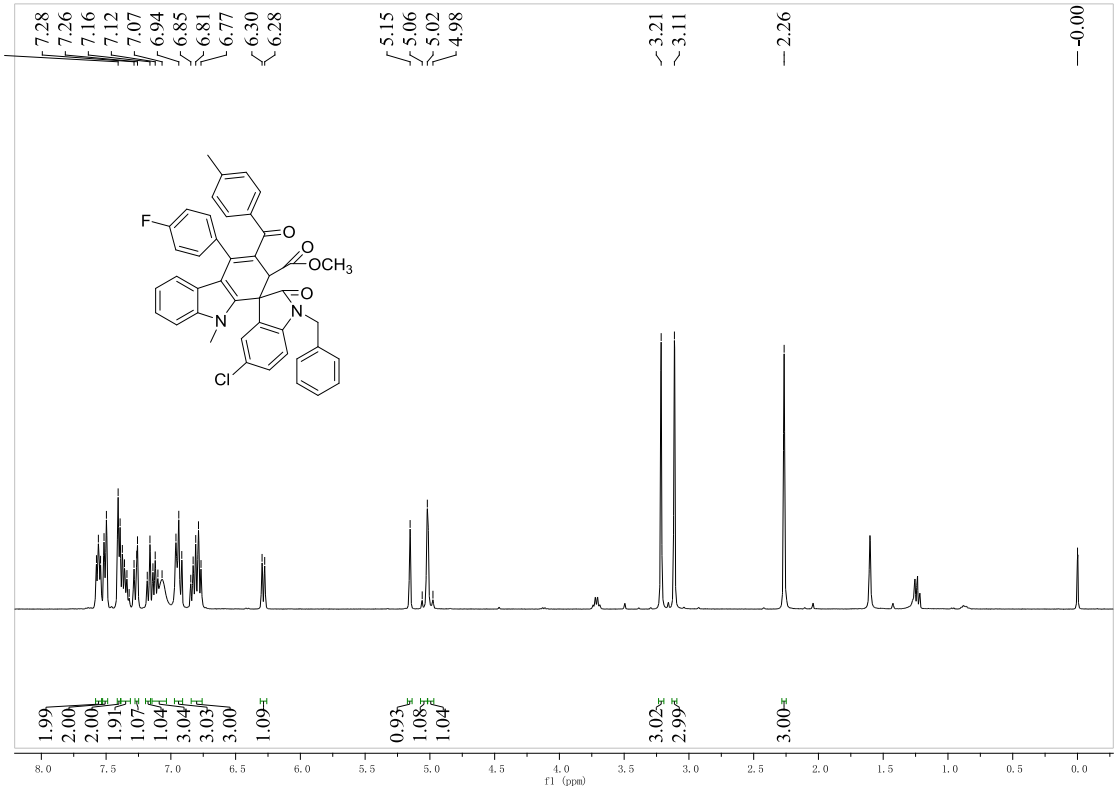

198.20
176.71
170.02
131.27
129.77
128.68
128.49
128.16
124.43
122.66
120.64
114.55
114.33
112.56
110.60
109.47
57.21
52.54
51.87
45.22
30.42
21.22
f1 (ppm)

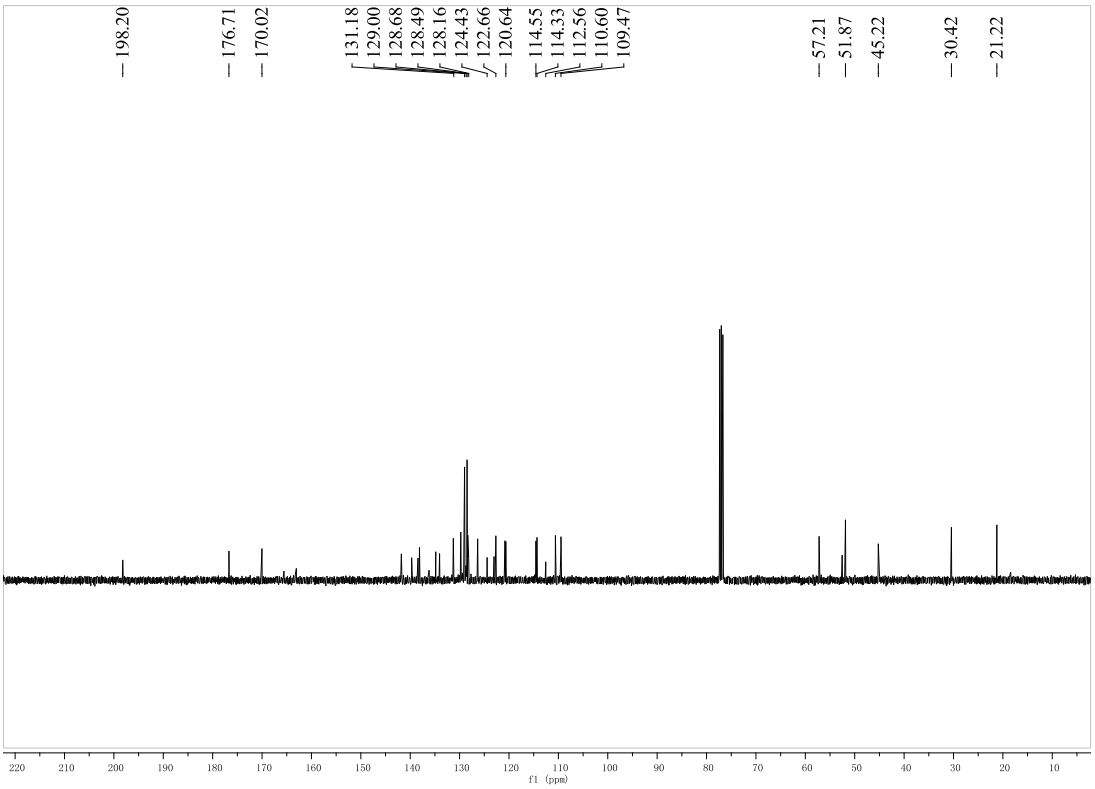

Intens.
x10^4
+MS, 0.14-0.17min #(8-10)
731.2110
732.2139
733.2100
734.2123
729.2033
735.2146
737.2835
m/z

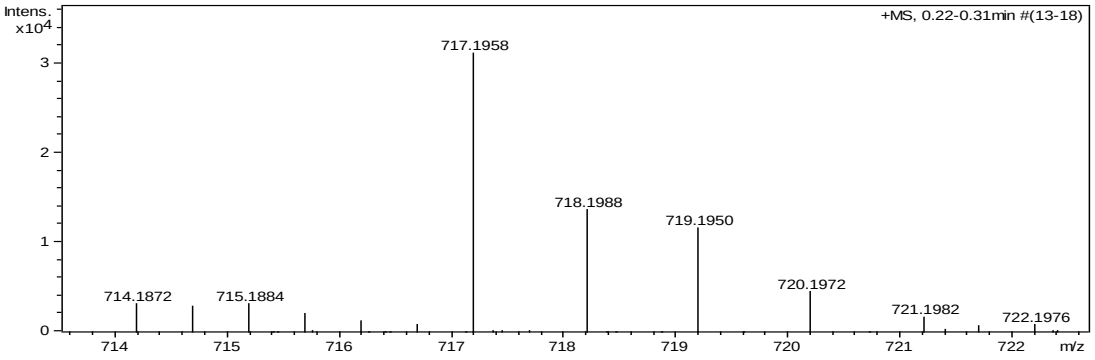
**Methyl**

**1'-benzyl-5'-chloro-4-(4-fluorophenyl)-9-methyl-3-(4-methylbenzoyl)-2'-oxo-2,9-dihydrospiro[carbazole-1,3'-indoline]-2-carboxylate (3m):**

white solid, 301 mg, 84%, m.p. 184-187 $^{o}$C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.58-7.54 (m, 2H, ArH), 7.52-7.50 (m, 2H, ArH), 7.41-7.34 (m, 4H, ArH), 7.29-7.26 (m, 1H, ArH), 7.17 (d, *J* = 8.0 Hz, 1H, ArH), 7.14-7.07 (m, 3H, ArH), 7.14-7.07 (m, 3H, ArH), 6.96-6.92 (m, 3H, ArH), 6.85-6.78 (m, 3H, ArH), 6.29 (d, *J* = 8.0 Hz, 1H, ArH), 5.15 (s, 1H, CH), 5.04 (d, *J* = 16.0 Hz, 1H, CH), 5.00 (d, *J* = 16.0 Hz, 1H, CH), 3.22 (s, 3H, $OCH_3$), 3.11 (s, 3H, $CH_3$), 2.27 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 198.2, 176.7, 170.0, 141.7, 139.7, 138.4, 138.1, 134.8, 134.0, 134.0, 131.2, 131.1, 129.7, 128.9, 128.7, 128.6, 128.6, 128.4, 128.3, 128.1, 126.3, 124.4, 123.0, 122.6, 120.8, 120.6, 114.5, 114.3, 112.5, 110.6, 109.4, 57.2, 52.5, 51.8, 45.2, 30.4, 21.2; IR(KBr) υ: 3189, 3061, 2949, 1910, 1729, 1603, 1508, 1472, 1324, 1276, 1227, 1183, 1121, 1083, 1017, 940, 806, 747, 701, 583 cm$^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{43}H_{32}ClFN_2O_4$ ([M+Na]$^{+}$): 717.1927, found:717.1958.

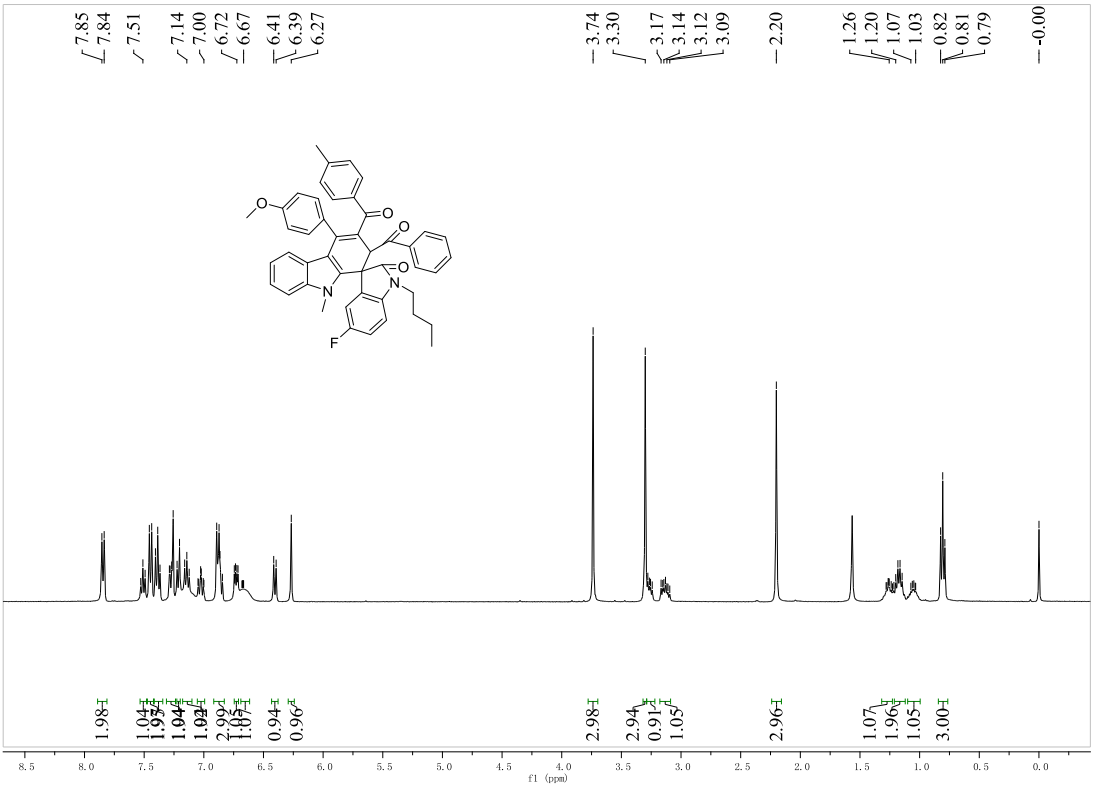


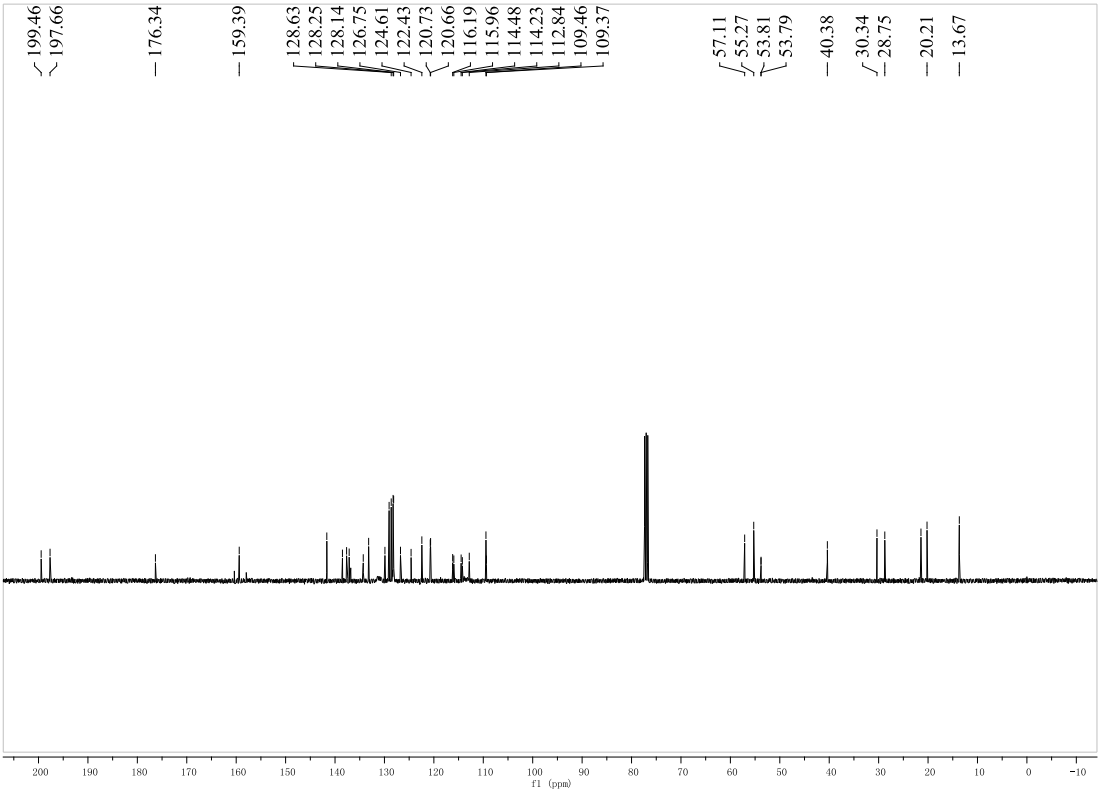

198.20
176.71
170.02
131.18
129.00
128.68
128.49
128.16
124.43
122.66
120.64
114.55
114.33
112.56
110.60
109.47
57.21
51.87
45.22
30.42
21.22
f1 (ppm)

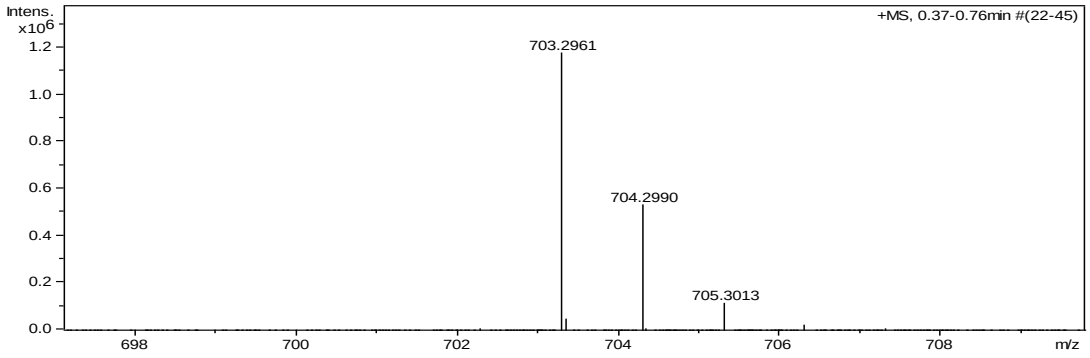

Intens.
x10^4
+MS, 0.22-0.31min #(13-18)
717.1958
718.1988
719.1950
720.1972
721.1982
722.1976
714.1872
715.1884
m/z

**(1'-Butyl-5'-fluoro-4-(4-methoxyphenyl)-9-methyl-2'-oxo-2,9-dihydrospiro[carbazole-1,3'-indoline]-2,3-diyl)bis(*p*-tolylmethanone) (3n):**

Yellow solid, 218 mg, 62%, m.p. 177-180 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.84 (d, *J* = 8.0 Hz, 2H, ArH), 7.53-7.49 (m, 1H, ArH), 7.47 (d, *J* = 8.0 Hz, 2H, ArH), 7.41-7.37 (m, 2H, ArH), 7.29-7.26 (m, 3H, ArH), 7.21 (d, *J* = 8.0 Hz, 1H, ArH), 7.16-7.12 (m, 2H, ArH), 7.05-7.00 (m, 1H, ArH), 6.89-6.84 (m, 3H, ArH), 6.75-6.71 (m, 1H, ArH), 6.68-6.67 (m, 1H, ArH), 6.40 (d, *J* = 8.0 Hz, 1H, ArH), 6.27 (s, 1H, CH), 3.74 (s, 3H, $OCH_3$), 3.30 (s, 3H, $CH_3$), 3.28-3.24 (m, 1H, CH), 3.17-3.10 (m, 1H, CH), 2.20 (s, 3H, $CH_3$), 1.28-1.24 (m, 1H, CH), 1.22-1.15 (m, 2H, $CH_2$), 1.07-1.03 (m, 1H, CH), 0.81 (d, *J* = 7.2 Hz, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 199.4, 197.6, 176.3, 159.3, 141.6, 138.4, 137.6, 137.1, 133.1, 129.9, 129.0, 128.6, 128.2, 126.7, 122.4, 120.7, 116.1, 115.9, 114.4, 114.2, 112.8, 109.4, 109.3, 57.1, 55.2, 53.8, 53.7, 40.3, 30.3, 28.7, 21.4, 20.2, 13.6; IR(KBr) υ: 3073, 2941, 1781, 1655, 1567, 1531, 1470, 1367, 1249, 1233, 1182, 1111, 1063, 855, 831, 749, 654 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{46}H_{39}FN_2O_4$ ($[M+H]^+$): 703.2967, found: 703.2961.

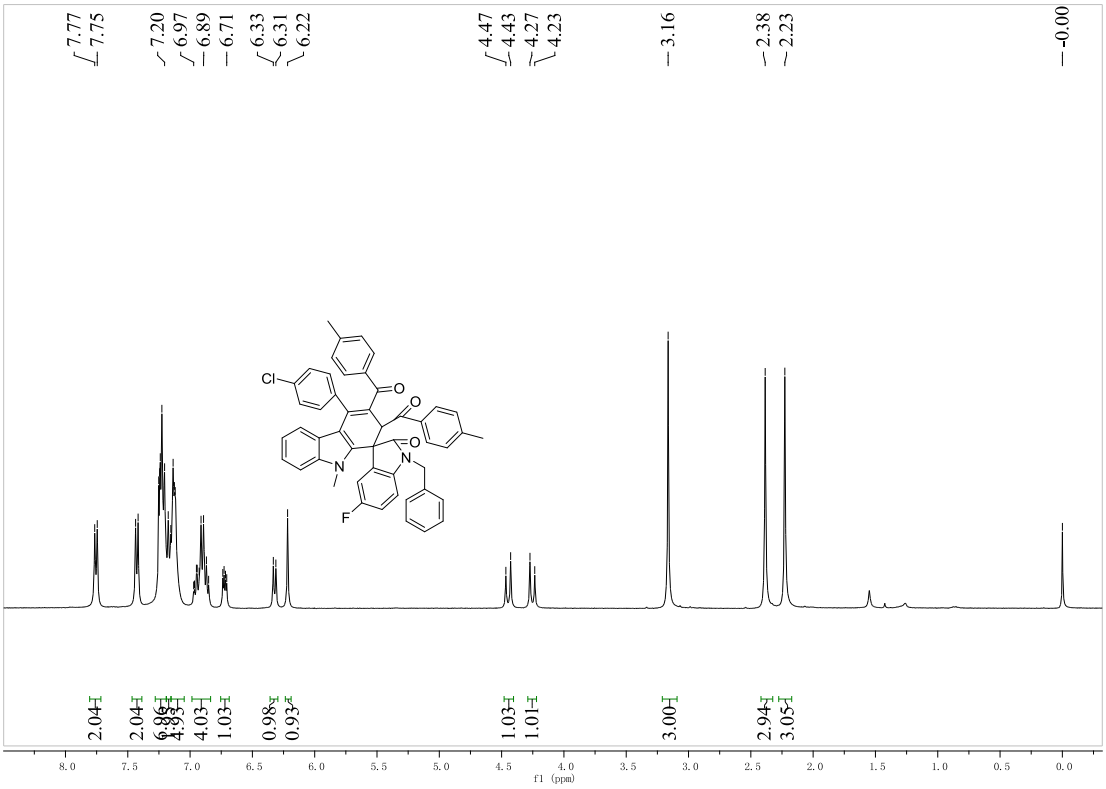


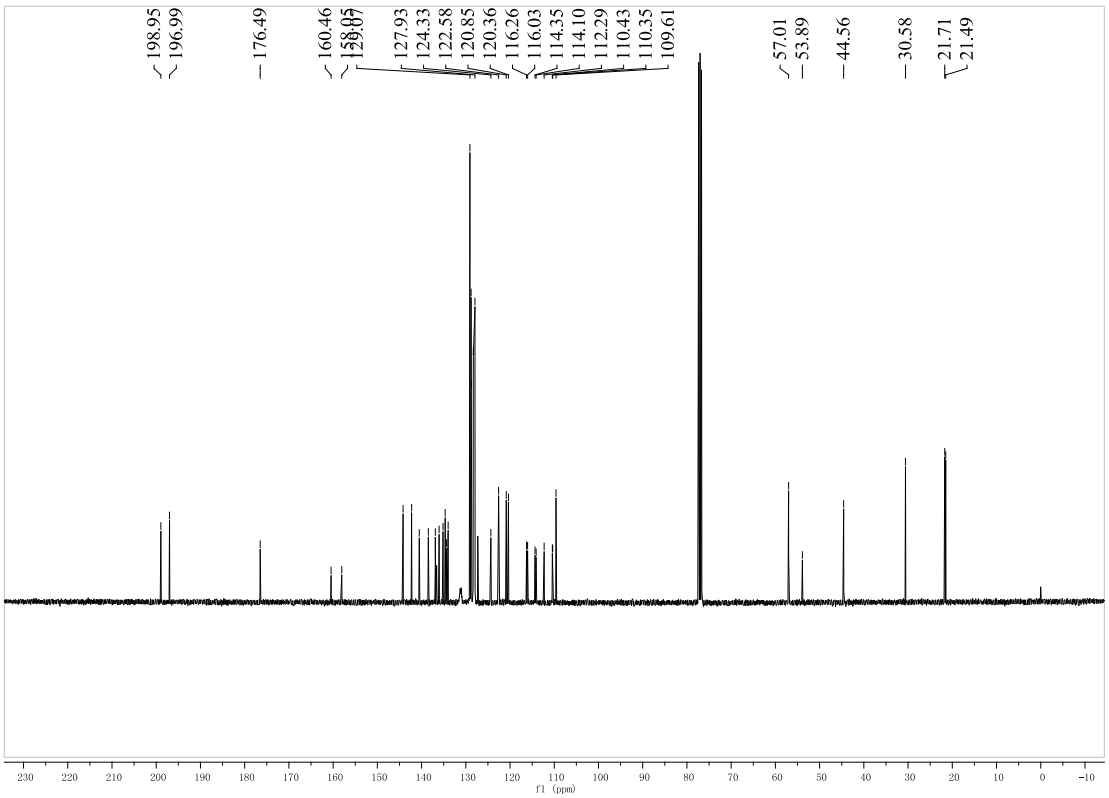

199.46
197.66
176.34
159.39
128.63
128.25
128.14
126.75
124.61
122.43
120.73
120.66
116.19
115.96
114.48
114.23
112.84
109.46
109.37
57.11
55.27
53.81
53.79
40.38
30.34
28.75
20.21
13.67
f1 (ppm)

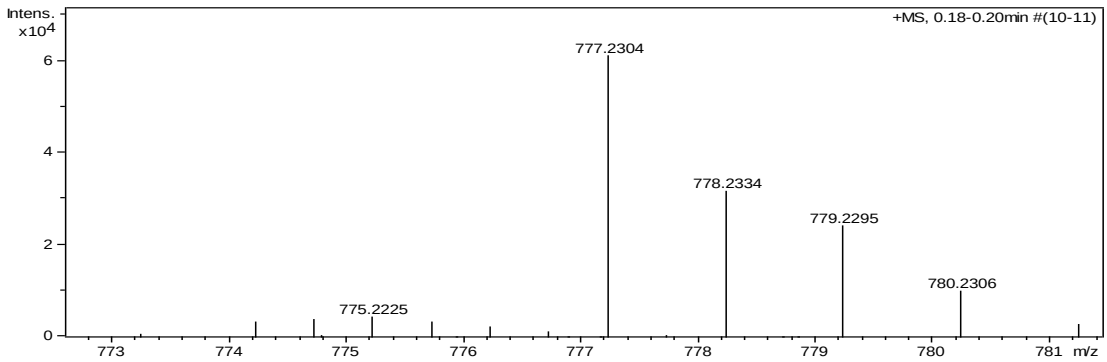

Intens.
x10^6
+MS, 0.37-0.76min #(22-45)
703.2961
704.2990
705.3013
m/z

**(1'-Benzyl-4-(4-chlorophenyl)-5'-fluoro-9-methyl-2'-oxo-2,9-dihydrospiro[carbazole-1,3'-indoline]-2,3-diyl)bis(*p*-tolylmethanone) (3o):**

Yellow solid, 303 mg, 78%, m.p. 168-172 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.74 (d, *J* = 8.0 Hz, 2H, ArH), 7.43 (d, *J* = 8.0 Hz, 2H, ArH), 7.25-7.21 (m, 7H, ArH), 7.18-7.16 (m, 2H, ArH), 7.14-7.12 (m, 5H, ArH), 6.97-6.85 (m, 4H, ArH), 6.74-6.71 (m, 1H, ArH), 6.32 (d, *J* = 8.0 Hz, 1H, ArH), 6.22 (s, 1H, CH), 4.45 (d, *J* = 15.2 Hz, 1H, CH), 4.25 (d, *J* = 15.2 Hz, 1H, CH), 3.16 (s, 3H, $CH_3$), 2.39 (s, 3H, $CH_3$), 2.23 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 198.9, 196.9, 176.4, 160.4, 158.0, 144.1, 142.2, 140.5, 138.4, 136.8, 136.0, 135.1, 134.6, 133.9, 129.0, 128.8, 127.9, 124.3, 122.5, 120.8, 120.3, 116.2, 114.1, 112.2, 110.4, 109.6, 57.0, 53.8, 44.5, 30.5, 21.7, 21.4; IR(KBr) υ: 3079, 3050, 1905, 1714, 1673, 1604, 1519, 1482, 1409, 1321, 1267, 1090, 953, 811, 740, 699, 609, 561 cm$^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{49}H_{36}ClFN_2O_3$ ([M+Na]$^{+}$): 777.2291, found: 777.2304.

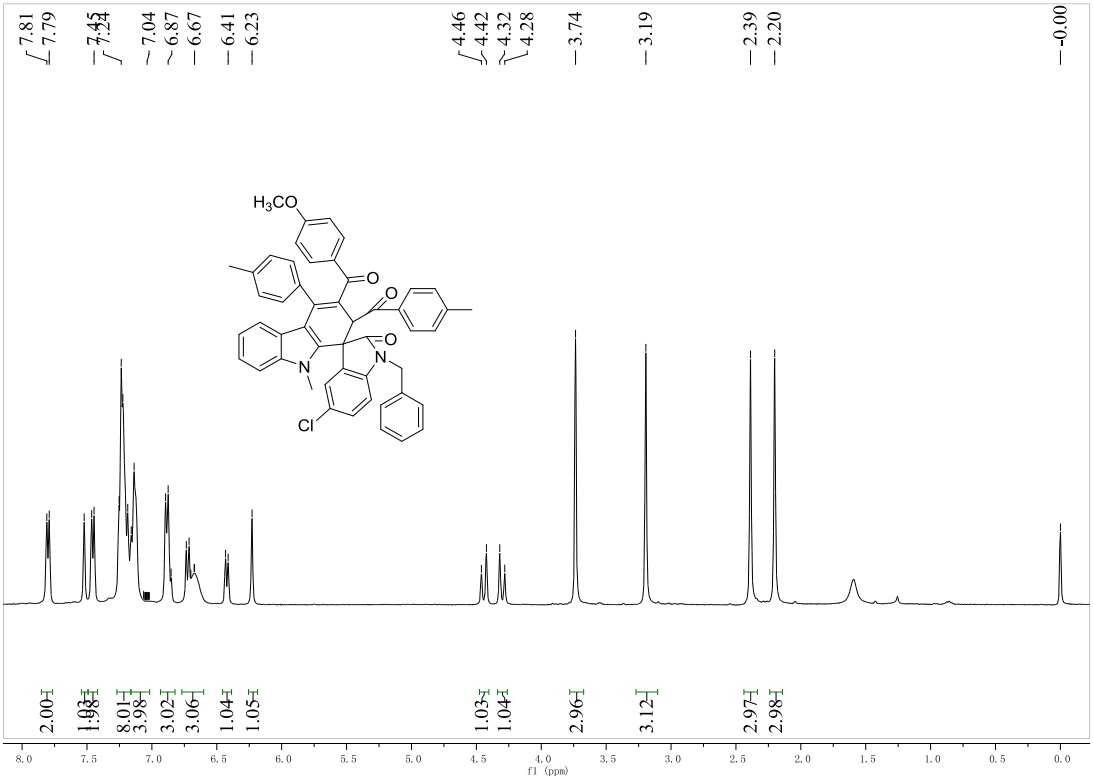


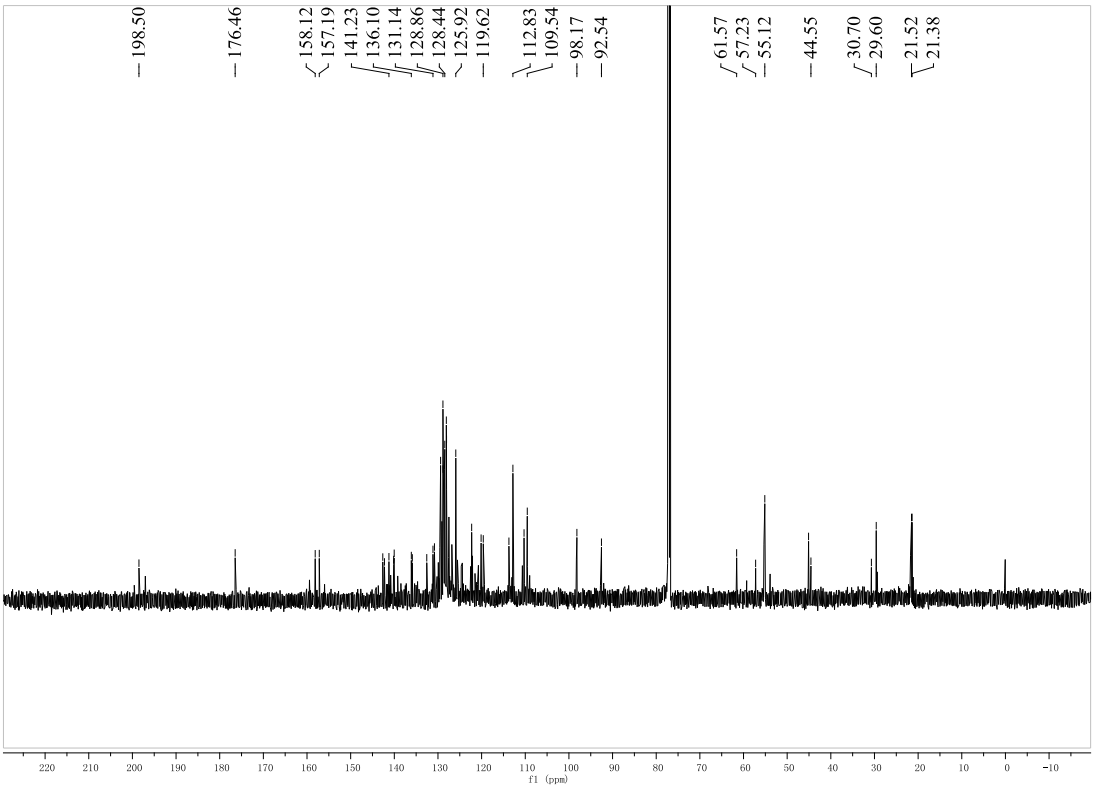

198.95
196.99
176.49
160.46
158.05
129.07
127.93
124.33
122.58
120.85
120.36
116.26
116.03
114.35
114.10
112.29
110.43
110.35
109.61
57.01
53.89
44.56
30.58
21.71
21.49
f1 (ppm)

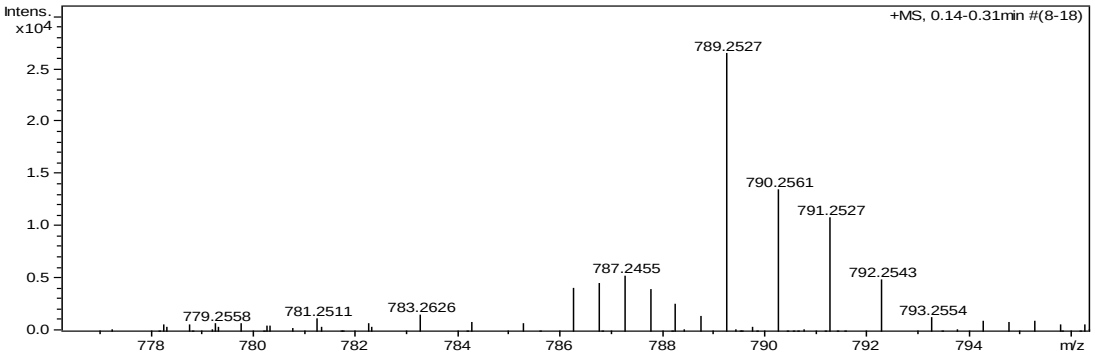

Intens.
x10^4
+MS, 0.18-0.20min #(10-11)
777.2304
778.2334
779.2295
780.2306
775.2225
m/z

**1'-Benzyl-5'-chloro-3-(4-methoxybenzoyl)-9-methyl-2-(4-methylbenzoyl)-4-(*p*-tolyl)-2,9-dihydrospiro[carbazole-1,3'-indolin]-2'-one (3p):**

yellow solid, 316 mg, 80%, m.p. 176-179 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.80 (d, *J* = 8.0 Hz, 2H, ArH), 7.52 (s, 1H, ArH), 7.46 (d, *J* = 8.0 Hz, 2H, ArH), 7.25-7.18 (m, 8H, ArH), 7.16-7.02 (m, 4H, ArH), 6.90-6.85 (m, 3H, ArH), 6.74-6.67 (m, 3H, ArH), 6.42 (d, *J* = 8.0 Hz, 1H, ArH), 6.23 (s, 1H, CH), 4.44 (d, *J* = 15.2 Hz, 1H, CH), 4.30 (d, *J* = 15.2 Hz, 1H, CH), 3.74 (s, 3H, $OCH_3$), 3.19 (s, 3H, $CH_3$), 2.39 (s, 3H, $CH_3$), 2.20 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 198.4, 176.4, 158.1, 157.1, 142.6, 142.2, 140.1, 136.1, 135.8, 132.5, 131.1, 129.3, 128.8, 128.5, 128.0, 125.9, 122.2, 120.1, 113.7, 112.8, 110.2, 109.5, 98.1, 61.5, 55.1, 45.1, 30.7, 29.5, 21.5; IR(KBr) υ: 3072, 2946, 1892, 1654, 1542, 1508, 1452, 1329, 1245, 1154, 1102, 1073, 937, 799, 725, 669, 557 $cm^{-1}$;MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{50}H_{39}ClN_2O_4$([M+Na]$^+$): 789.2491, found:789.2527.

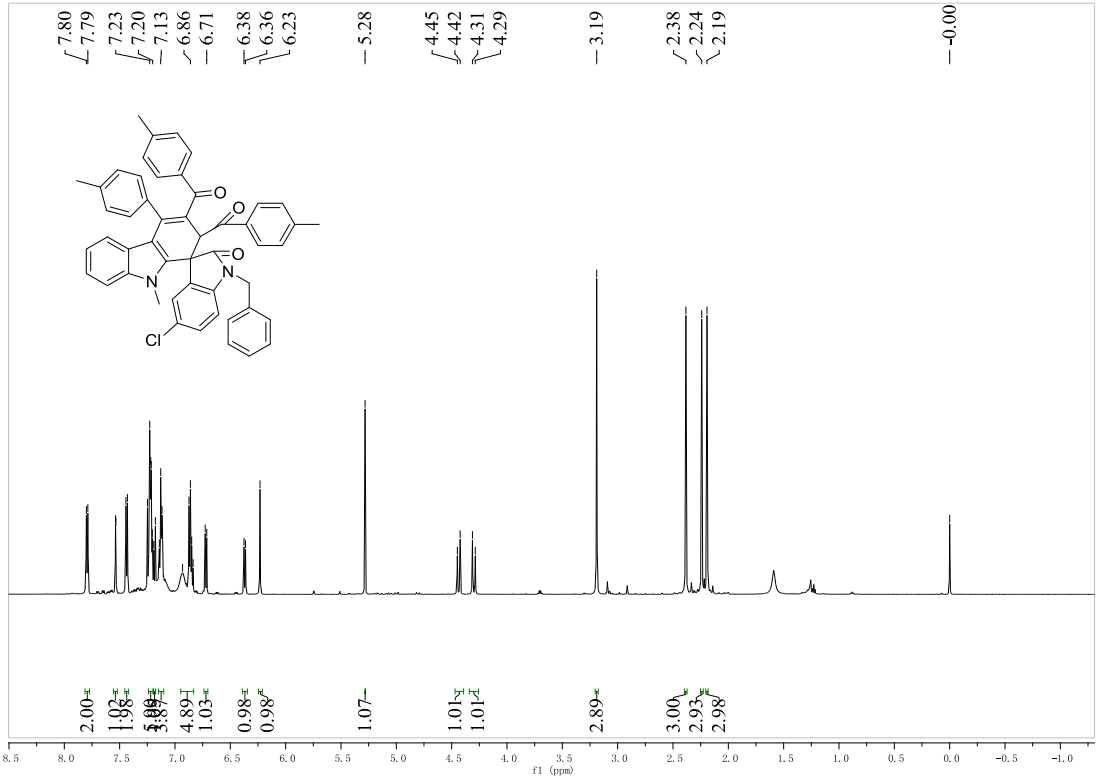


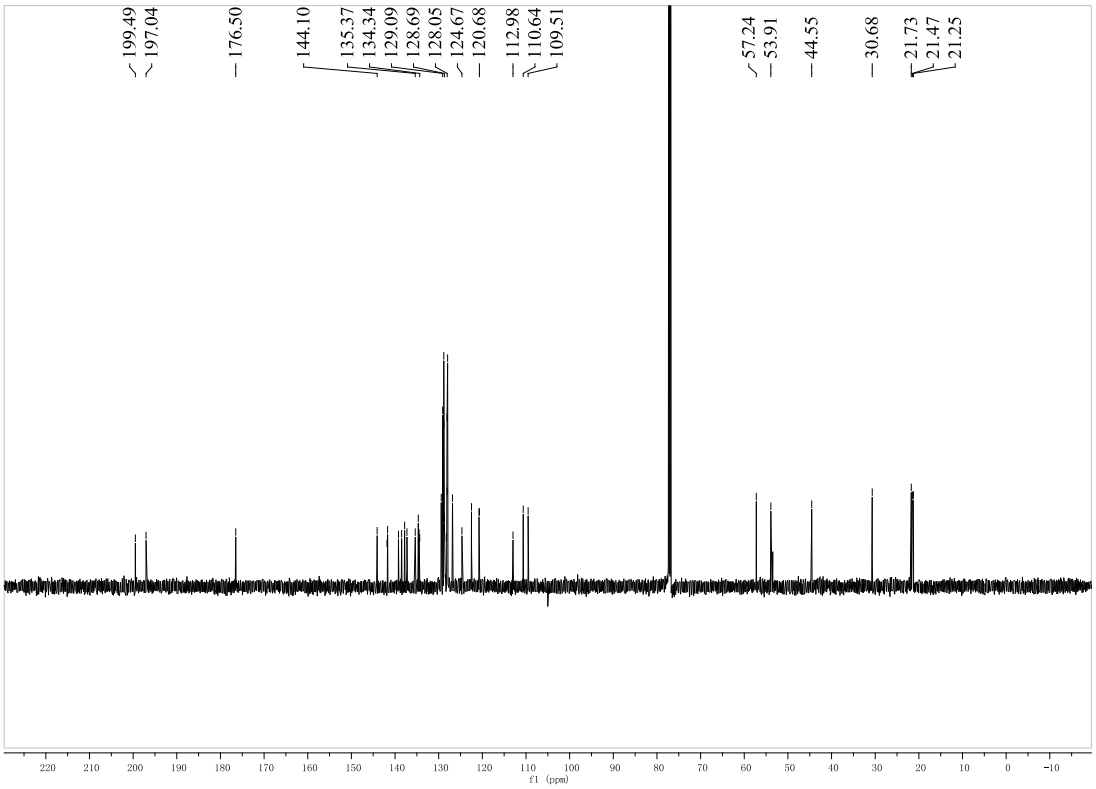

198.50
176.46
158.12
157.19
141.23
136.10
131.14
128.86
128.44
125.92
119.62
112.83
109.54
98.17
92.54
61.57
57.23
55.12
44.55
30.70
29.60
21.52
21.38
f1 (ppm)

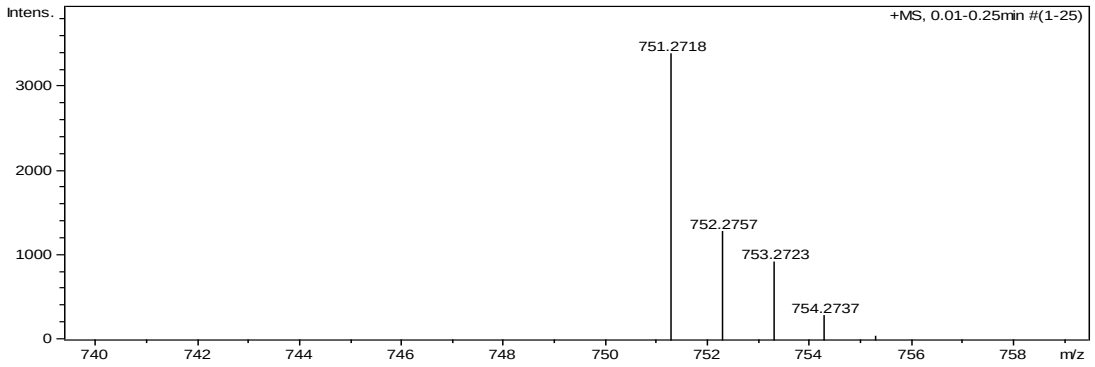

Intens.
x10^4
+MS, 0.14-0.31min #(8-18)
789.2527
790.2561
791.2527
792.2543
793.2554
787.2455
779.2558
781.2511
783.2626
m/z

**(1'-Benzyl-5'-chloro-9-methyl-2'-oxo-4-(p-tolyl)-2,9-dihydrospiro[carbazole-1,3'-indoline]-2, 3-diyl)bis(*p*-tolylmethanone) (3q):**

yellow solid, 316 mg, 84%, m.p. 181-184 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.79 (d, *J* = 5.6 Hz, 2H, ArH), 7.54-7.53 (m, 1H, ArH), 7.44 (d, *J* = 5.6 Hz, 2H, ArH), 7.23-7.20 (m, 6H, ArH), 7.18 (d, *J* = 5.6 Hz, 1H, ArH), 7.14-7.12 (m, 4H, ArH), 6.93-6.84 (m, 5H, ArH), 6.72 (d, *J* = 5.6 Hz, 1H, ArH), 6.37 (d, *J* = 5.6 Hz, 1H, ArH), 6.23 (s, 1H, ArH), 5.28 (s, 1H, CH), 4.44 (d, *J* = 10.4 Hz, 1H, CH), 4.30 (d, *J* = 10.4 Hz, 1H, CH), 3.19 (s, 3H, $CH_3$), 2.83 (s, 3H, $CH_3$), 2.24 (s, 3H, $CH_3$), 2.19 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 199.4, 197.0, 176.4, 144.1, 141.7, 139.2, 138.4, 137.8, 137.2, 135.3, 134.6, 129.4, 129.1, 129.0, 128.8, 128.6, 128.2, 128.0, 127.9, 126.8, 124.6, 122.4, 120.7, 120.6, 112.9, 110.6, 109.5, 57.2, 53.9, 44.5, 30.6, 21.7, 21.4, 21.2; IR (KBr) υ: 3022, 2930, 1852, 1633, 1532, 1511, 1452, 1306, 1245, 1126, 1102, 1033, 931, 759, 720, 639, 521 $cm^{-1}$;MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{50}H_{39}ClN_2O_4$ ($[M+H]^+$): 751.2722, found:751.2718.

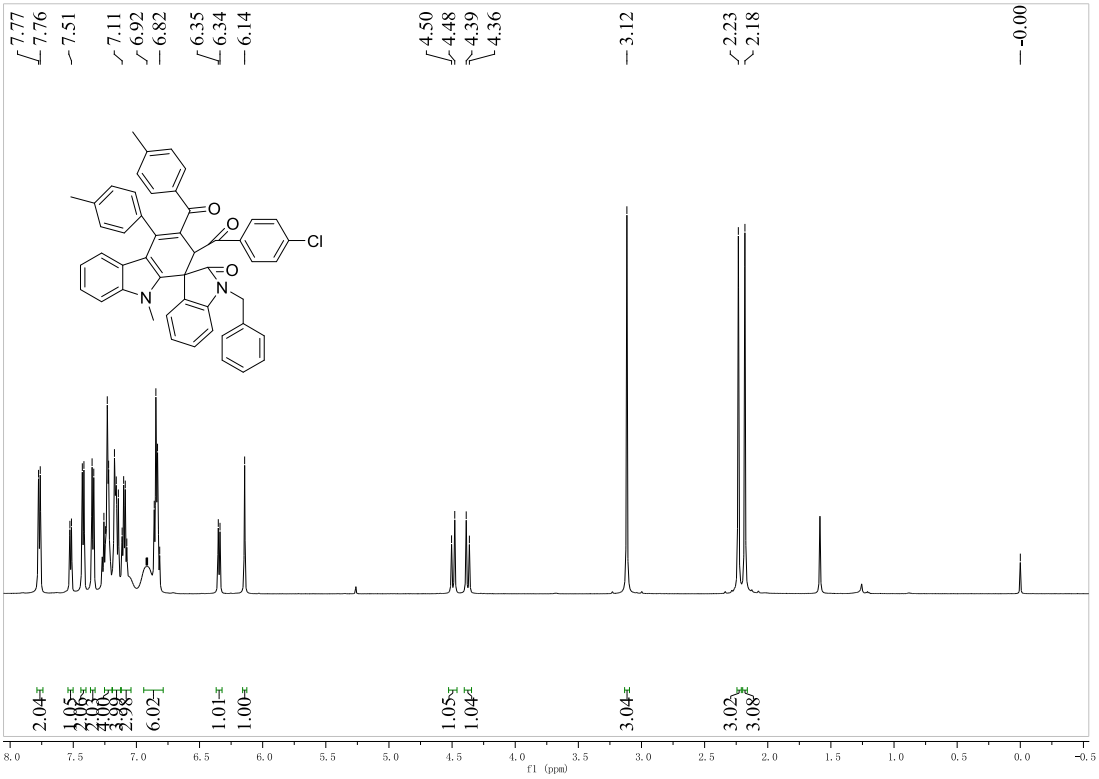


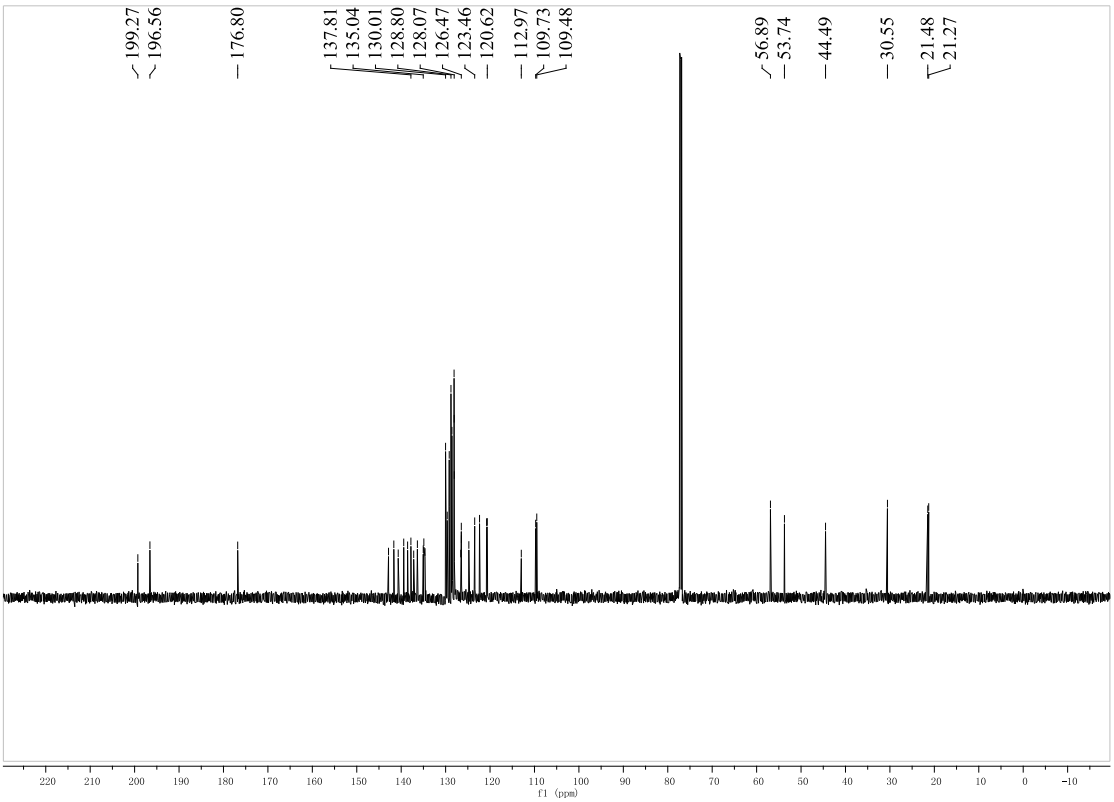

199.49
197.04
176.50
144.10
135.37
134.34
129.09
128.69
128.05
124.67
120.68
112.98
110.64
109.51
57.24
53.91
44.55
30.68
21.73
21.47
21.25
f1 (ppm)

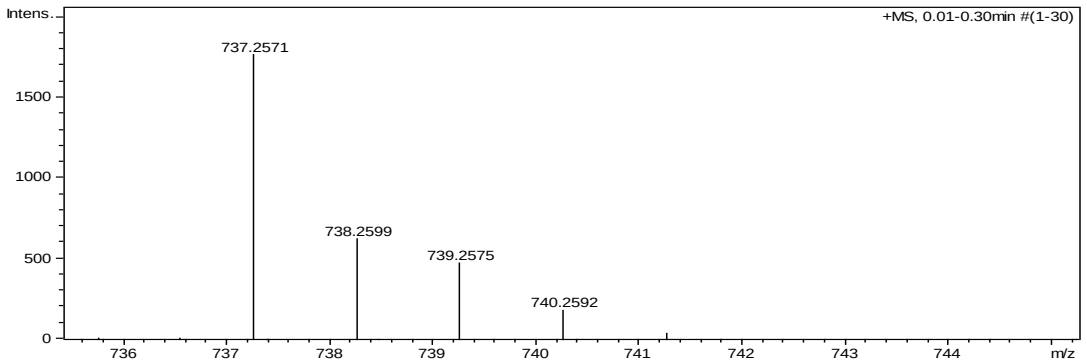

Intens.
+MS, 0.01-0.25min #(1-25)
751.2718
752.2757
753.2723
754.2737
m/z

**1'-Benzyl-2-(4-chlorobenzoyl)-9-methyl-3-(4-methylbenzoyl)-4-(*p*-tolyl)-2,9-dihydrospiro[carbazole-1,3'-indolin]-2'-one (3r):**

yellow solid, 328 mg, 89%, m.p. 165-167 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.77 (d, *J* = 5.2 Hz, 2H, ArH), 7.52 (d, *J* = 5.2 Hz, 1H, ArH), 7.42 (d, *J* = 5.2 Hz, 2H, ArH), 7.34 (d, *J* = 5.2 Hz, 2H, ArH), 7.24-7.22 (m, 4H, ArH), 7.17-7.14 (m, 4H, ArH), 7.11-7.08 (m, 3H, ArH), 6.92-6.82 (m, 6H, ArH), 6.34 (d, *J* = 5.2 Hz, 1H, ArH), 6.14 (s, 1H, CH), 4.49 (d, *J* = 10.0 Hz, 1H, CH), 4.37 (d, *J* = 10.0 Hz, 1H, CH), 3.12 (s, 3H, $CH_3$), 2.23 (s, 3H, $CH_3$), 2.18 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 199.2, 196.5, 176.7, 142.8, 141.6, 140.6, 139.4, 138.5, 137.8, 137.1, 136.3, 135.0, 134.9, 134.6, 130.0, 129.6, 129.1, 128.8, 128.6, 128.1, 128.0, 127.9, 126.5, 126.4, 126.3, 124.7, 123.4, 122.3, 120.7, 120.6, 112.9, 109.7, 109.4, 56.8, 53.7, 44.4, 30.5, 21.4, 21.2; IR (KBr) υ: 3032, 2917, 1822, 1603, 1532, 1500, 1407, 1346, 1215, 1176, 1102, 1063, 933, 789, 710, 635, 531 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{49}H_{37}ClN_2O_3$ ($[M+H]^+$): 737.2565, found:737.2571.

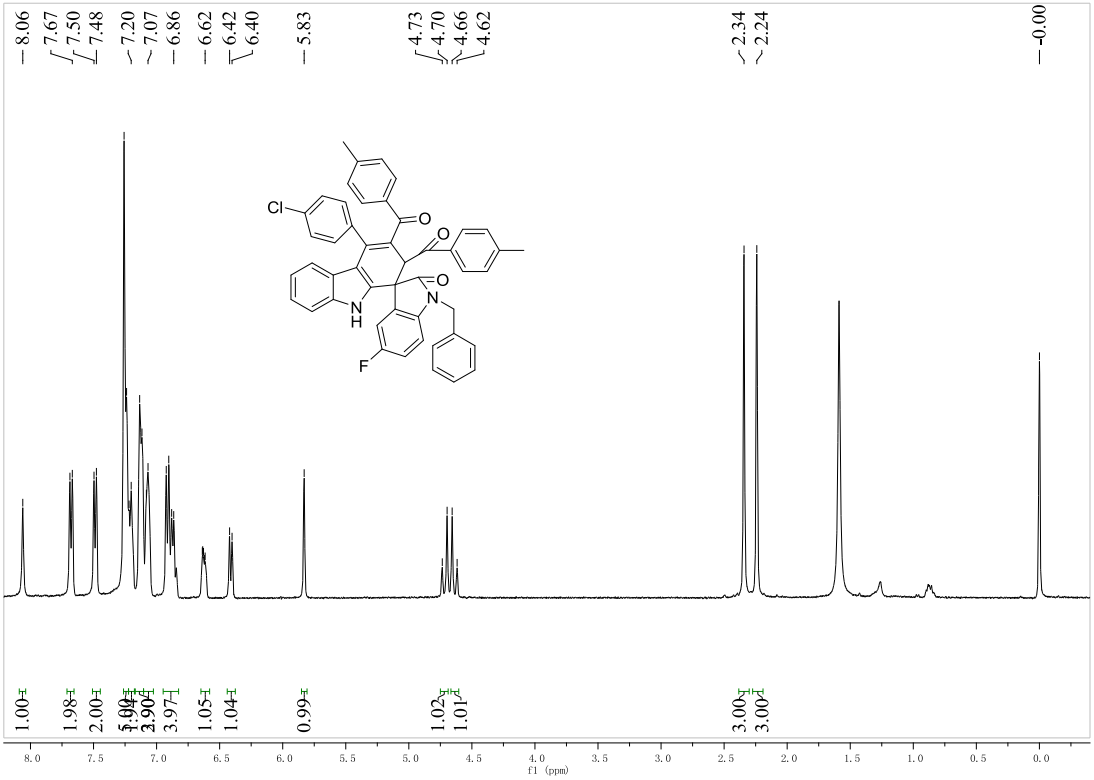


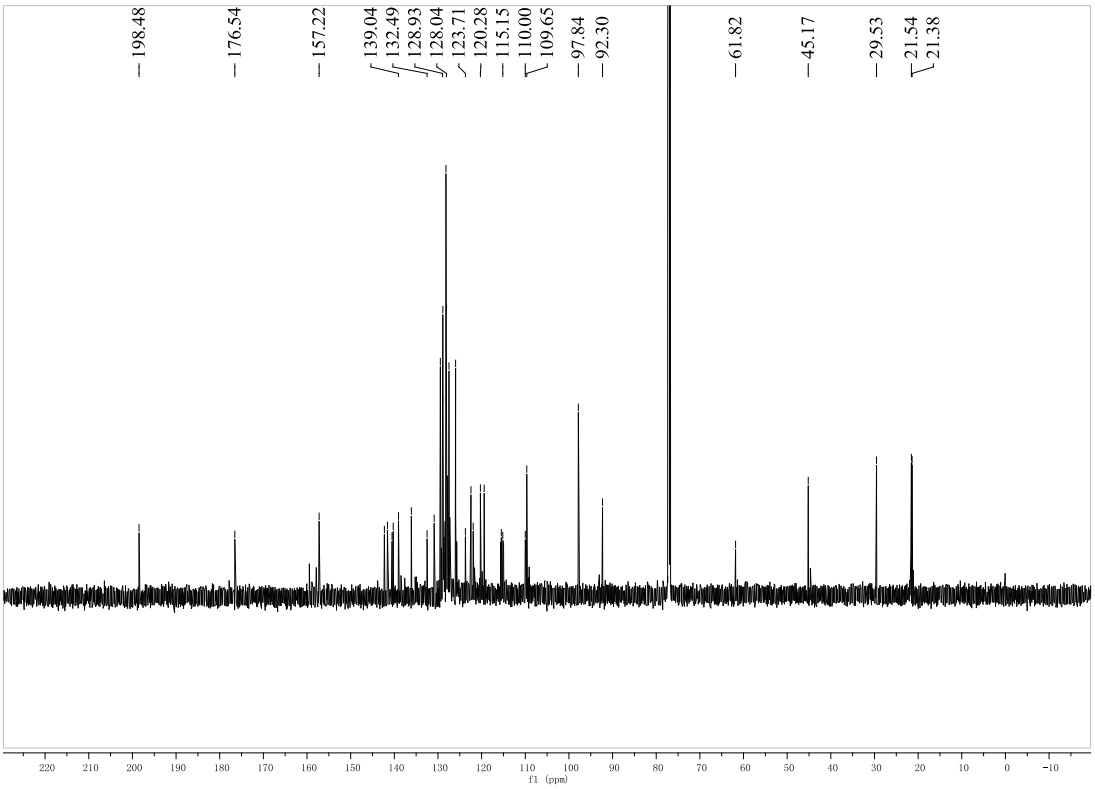

199.27
196.56
176.80
137.81
135.04
130.01
128.80
128.07
126.47
123.46
120.62
112.97
109.73
109.48
56.89
53.74
44.49
30.55
21.48
21.27
f1 (ppm)

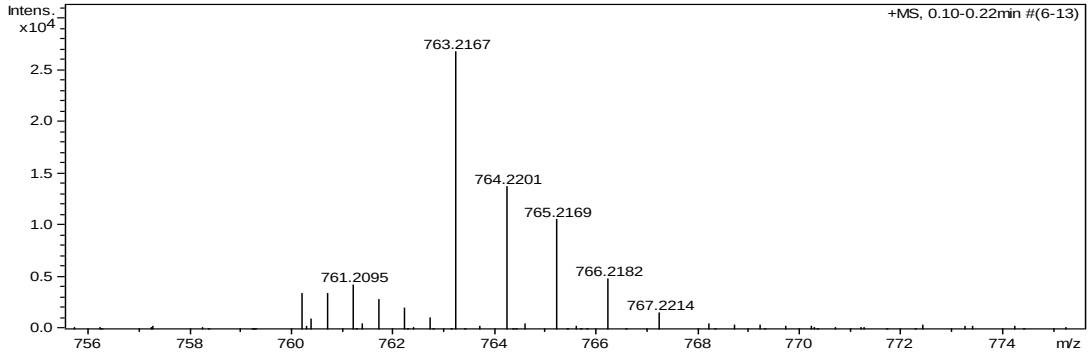

Intens.
+MS, 0.01-0.30min #(1-30)
737.2571
738.2599
739.2575
740.2592
m/z

**(1'-Benzyl-4-(4-chlorophenyl)-5'-fluoro-2'-oxo-2,9-dihydrospiro[carbazole-1,3'-indoline]-2,3-diyl)bis(*p*-tolylmethanone) (3s):**

Yellow solid, 294 mg, 77%, m.p. 171-174 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 8.06 (s, 1H, NH), 7.68 (d, *J* = 8.0 Hz, 2H, ArH), 7.49 (d, *J* = 8.0 Hz, 2H, ArH), 7.26-7.24 (m, 5H, ArH), 7.22-7.20 (m, 2H, ArH), 7.13-7.11 (m, 4H, ArH), 7.08-7.07 (m, 3H, ArH), 6.92-6.86 (m, 4H, ArH), 6.64-6.62 (m, 1H, ArH), 6.41 (d, *J* = 8.0 Hz, 1H, ArH), 5.83 (s, 1H, CH), 4.72 (d, *J* = 8.0 Hz, 1H, CH), 4.64 (d, *J* = 8.0 Hz, 1H, CH), 2.34 (s, 3H, $CH_3$), 2.24 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 198.4, 176.5, 157.2, 142.2, 141.5, 140.5, 139.0, 136.0, 132.4, 130.8, 129.4, 128.9, 128.8, 128.0, 127.4, 125.9, 123.7, 122.4, 121.9, 120.2, 119.4, 115.4, 115.1, 109.9, 97.8, 61.8, 45.1, 29.5, 21.5; IR (KBr) υ: 3017, 2906, 1957, 1649, 1583, 1520, 1458, 1300, 1245, 1124, 1105, 1014, 930, 839, 720, 676, 550 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{48}H_{34}ClFN_2O_3$ ($[M+Na]^+$): 763.2134, found:763.2167.

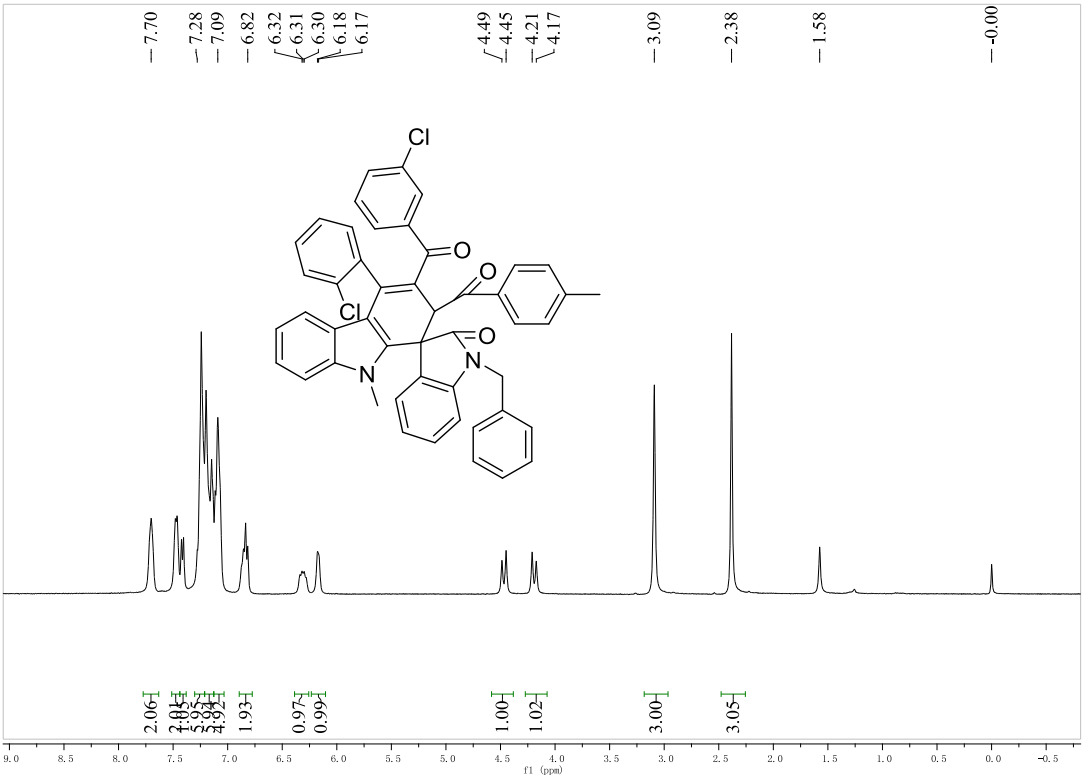


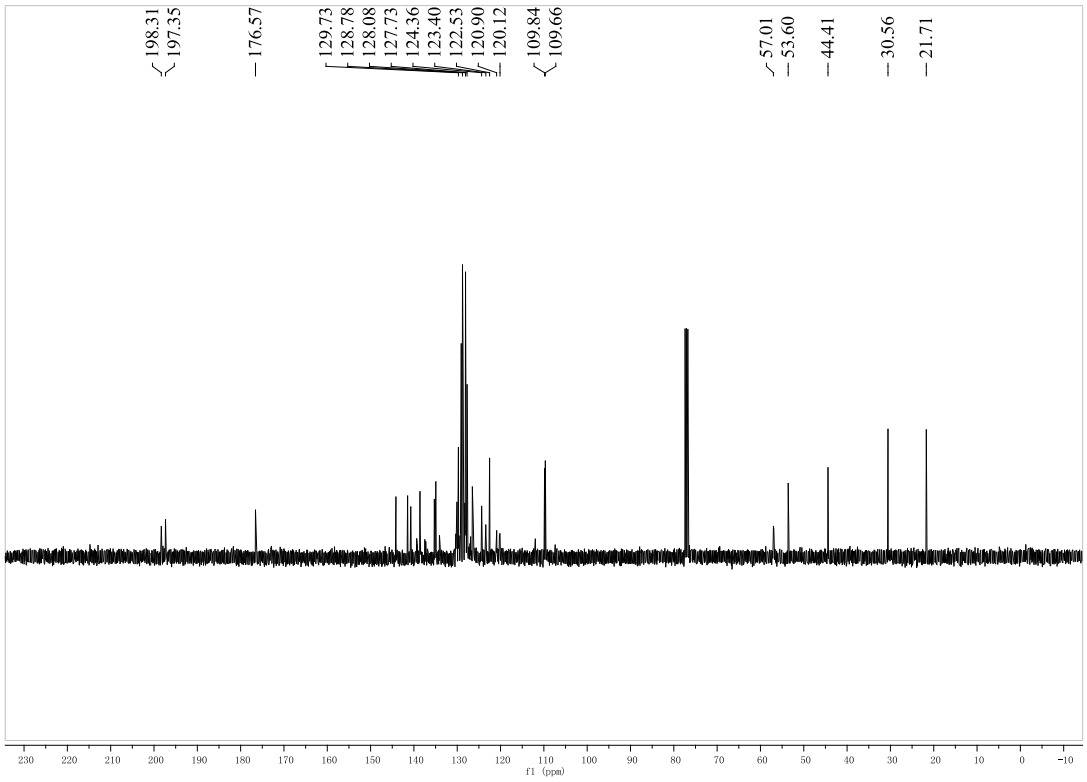

198.48
176.54
157.22
139.04
132.49
128.93
128.04
123.71
120.28
115.15
110.00
109.65
97.84
92.30
61.82
45.17
29.53
21.54
21.38
f1 (ppm)

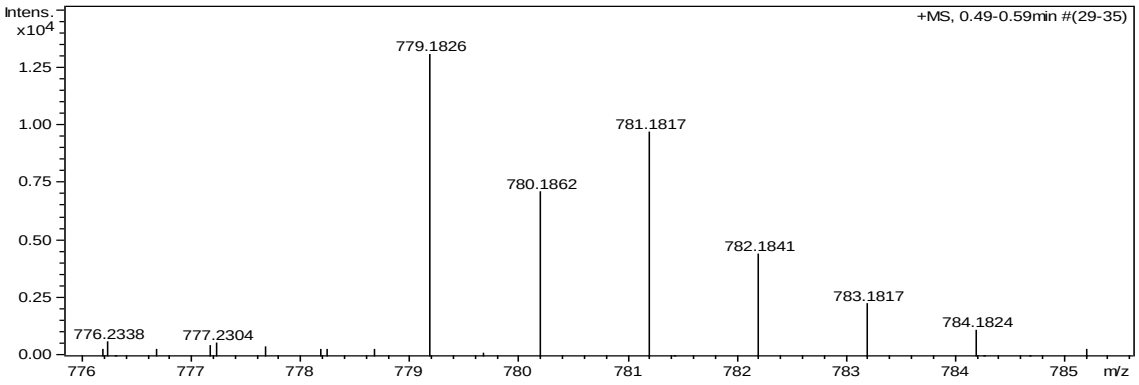

Intens.
x10^4
+MS, 0.10-0.22min #(6-13)
763.2167
764.2201
765.2169
766.2182
767.2214
761.2095
m/z

**1'-Benzyl-3-(3-chlorobenzoyl)-4-(2-chlorophenyl)-9-methyl-2-(4-methylbenzoyl)-2,9-dihydro spiro[carbazole-1,3'-indolin]-2'-one (3t):**

Yellow solid, 273 mg, 70%, m.p. 165-167 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.70-7.69 (m, 2H, ArH), 7.47 (d, *J* = 6.0 Hz, 2H, ArH), 7.41 (d, *J* = 7.2 Hz, 1H, ArH), 7.28-7.24 (m, 6H, ArH), 7.20-7.15 (m, 6H, ArH), 7.12-7.09 (m, 5H, ArH), 6.84 (t, *J* = 7.6 Hz, 2H, ArH), 6.32-6.29 (m, 1H, ArH), 6.17 (s, 1H, CH), 4.46 (d, *J* = 15.2 Hz, 1H, CH), 4.19 (d, *J* = 15.6 Hz, 1H, CH), 3.09 (s, 3H, $CH_3$), 2.38 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 198.3, 197.3, 176.5, 144.1, 140.7, 138.6, 134.9, 130.1, 129.7, 129.0, 128.7, 128.6, 128.0, 127.7, 126.5, 124.3, 123.4, 123.3, 122.5, 121.0, 120.9, 120.8, 120.1, 109.8, 109.6, 57.0, 53.6, 44.4, 30.5, 21.7; IR (KBr) υ: 3043, 2966, 1934, 1667, 1571, 1534, 1460, 1326, 1268, 1143, 1121, 1055, 989, 871, 763, 645, 563 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{48}H_{34}Cl_2N_2O_3$ ($[M+Na]^+$): 779.1839, found:779.1826.

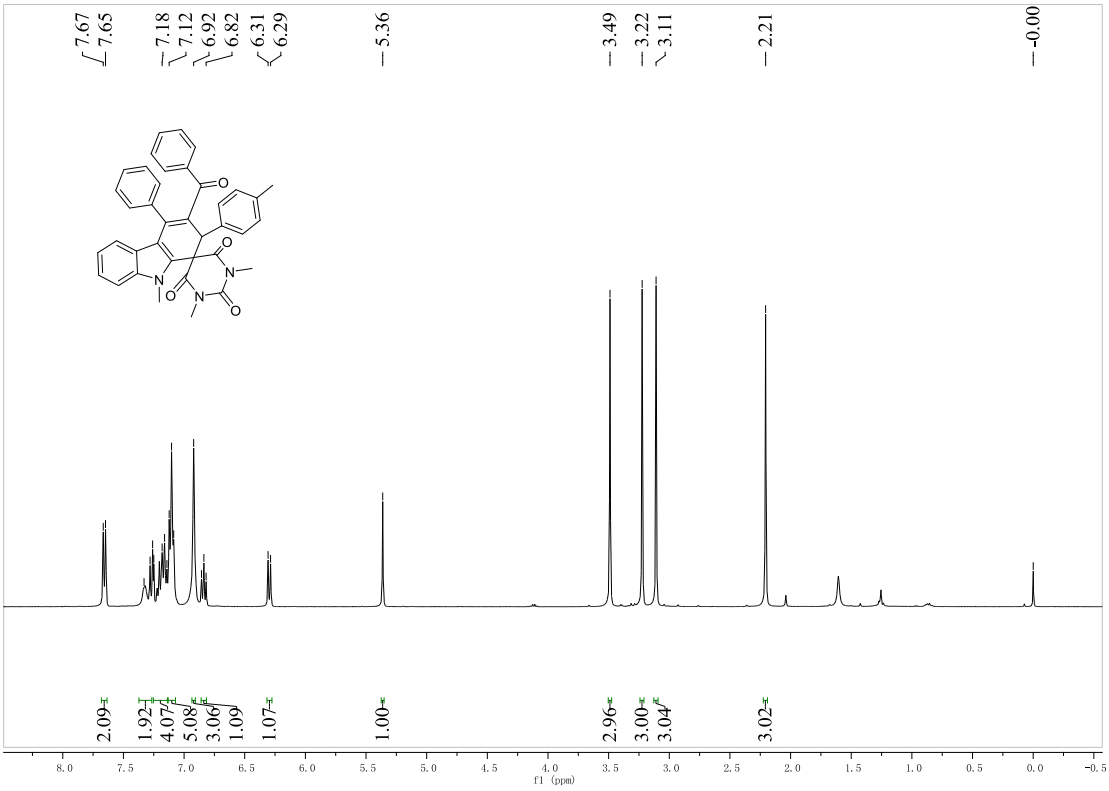


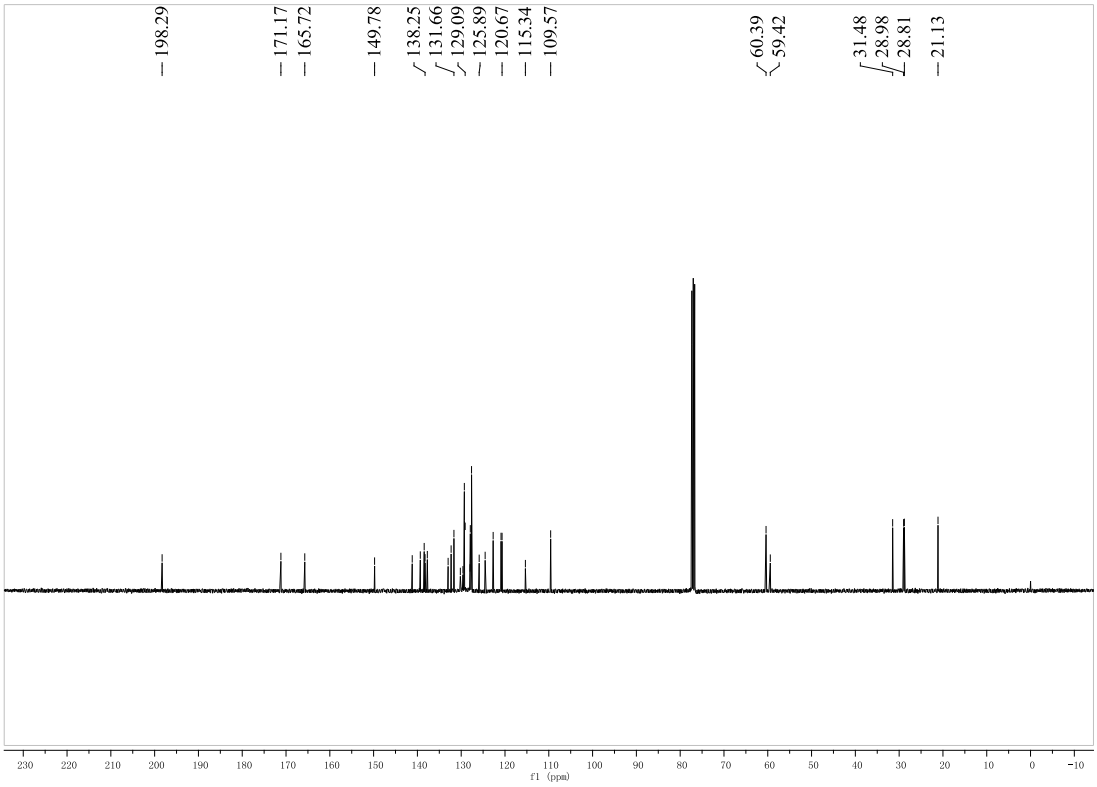

198.31
197.35
176.57
129.73
128.78
128.08
127.73
124.36
123.40
122.53
120.90
120.12
109.84
109.66
57.01
53.60
44.41
30.56
21.71
f1 (ppm)

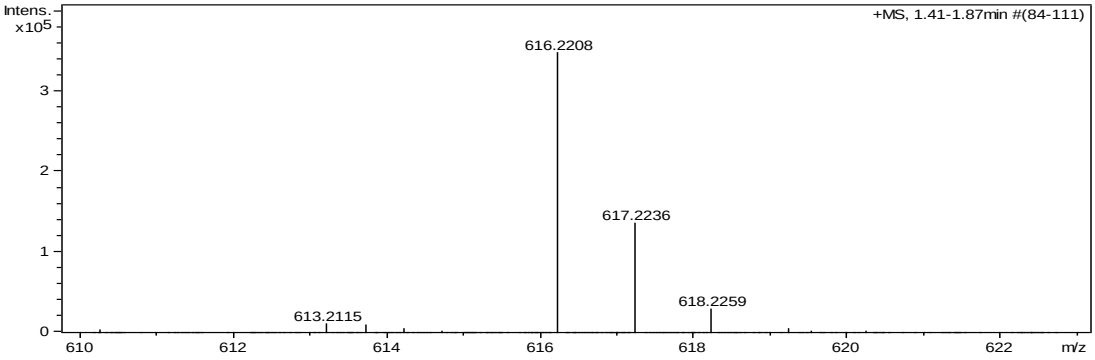

Intens.
x10^4
+MS, 0.49-0.59min #(29-35)
779.1826
781.1817
780.1862
782.1841
783.1817
784.1824
776.2338
777.2304
m/z

**3-Benzoyl-1',3',9-trimethyl-4-phenyl-2-(*p*-tolyl)-2,9-dihydro-2'H-spiro[carbazole-1,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (5a):**

Yellow solid, 241 mg, 78%, m.p. 184-187 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.66 (d, *J* = 7.2 Hz, 2H, ArH), 7.33-7.28 (m, 2H, ArH), 7.25-7.14 (m, 4H, ArH), 7.12-7.09 (m, 5H, ArH), 6.92 (s, 3H, ArH), 6.84(t, *J* = 7.2 Hz, 1H, ArH), 6.30 (d, *J* =8.4 Hz, 1H, ArH), 5.36 (s, 1H, CH), 3.49 (s, 3H, $CH_3$), 3.22 (s, 3H, $CH_3$), 3.11 (s, 3H, $CH_3$), 2.21 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 198.3, 171.2, 165.7, 149.8, 141.2, 139.3, 138.5, 138.3, 137.7, 133.0, 132.3, 131.7, 130.2, 129.6, 129.3, 129.1, 127.9, 127.8, 127.6, 125.9, 124.5, 122.7, 120.9, 120.7, 115.3, 109.6, 60.4, 59.4, 31.5, 29.0, 28.8, 21.1; IR (KBr) υ: 3133, 3040, 2913, 1931, 1755, 1621, 1574，1463, 1355, 1254, 1207, 1140, 1119, 1067, 1055, 990, 812, 754, 718 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{38}H_{31}N_3O_4$ ($[M+Na]^+$): 616,2207, found: 616,2208.

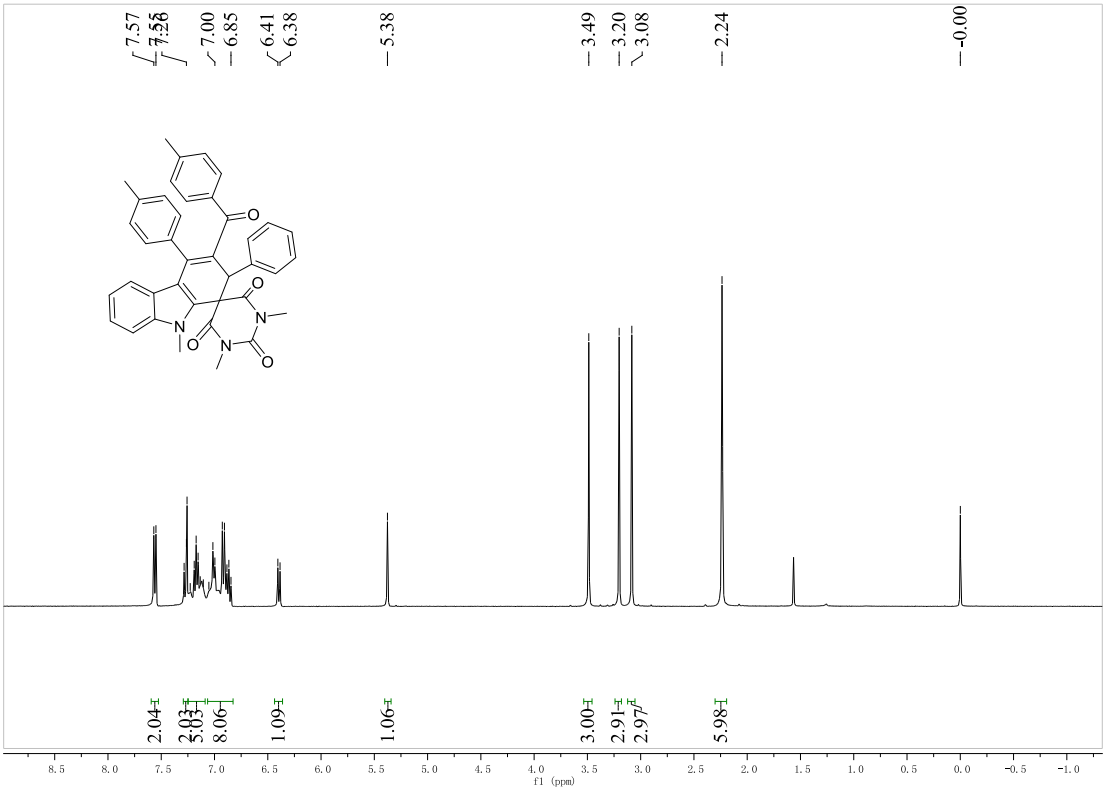


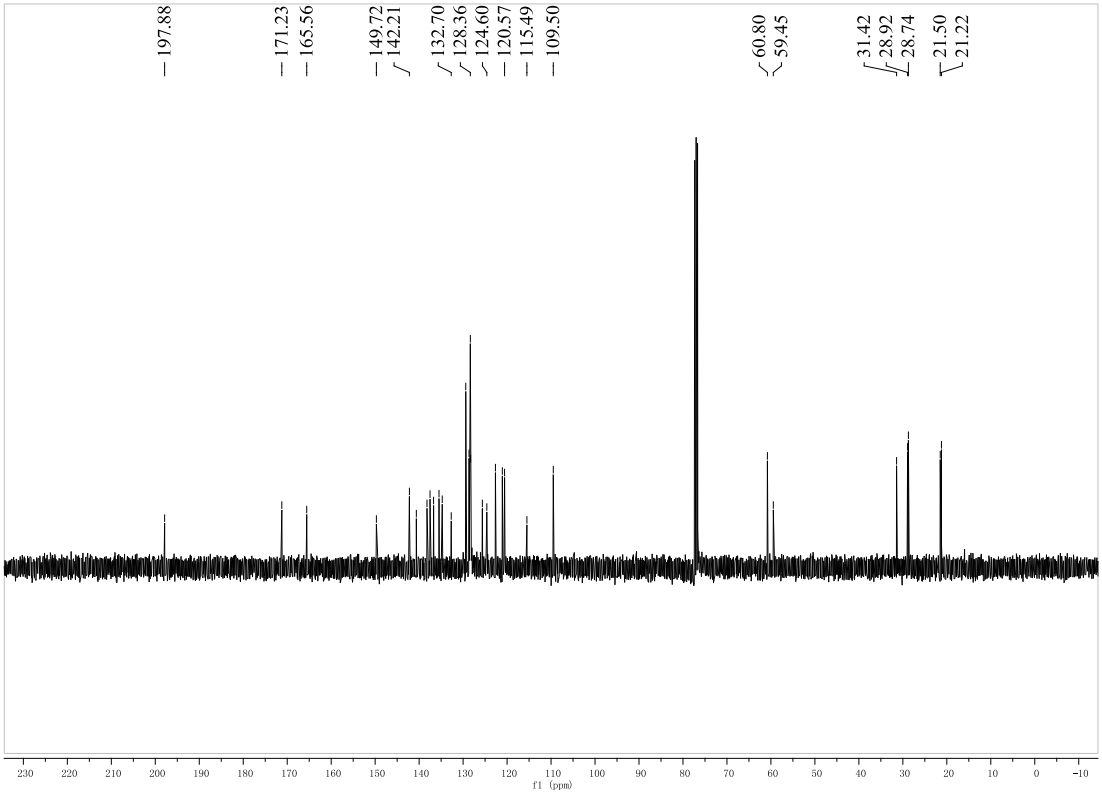

198.29
171.17
165.72
149.78
138.25
131.66
129.09
125.89
120.67
115.34
109.57
60.39
59.42
31.48
28.98
28.81
21.13
f1 (ppm)

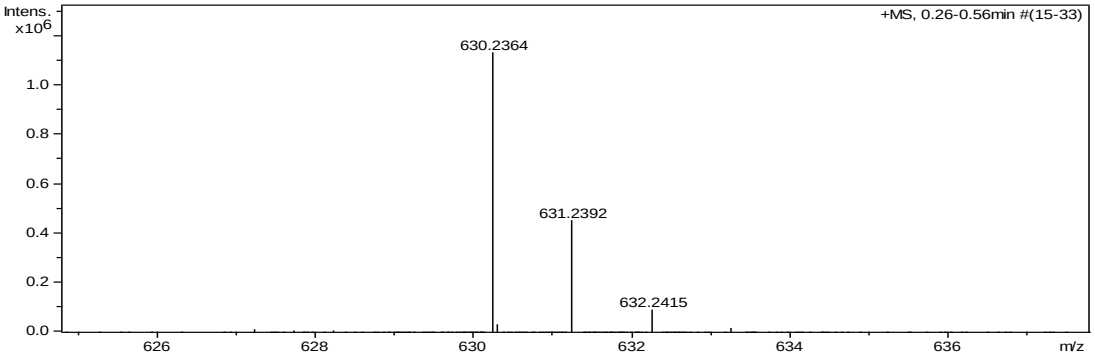

Intens.
x10^5
+MS, 1.41-1.87min #(84-111)
616.2208
617.2236
618.2259
613.2115
m/z

**1',3',9-Trimethyl-3-(4-methylbenzoyl)-2-phenyl-4-(*p*-tolyl)-2,9-dihydro-2'H-spiro[carbazole-1,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (5b):**

Yellow solid, 224 mg, 71%, m.p. 185-188 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.56 (d, *J* = 8.0 Hz, 2H, ArH), 7.27 (d, *J* = 8.8 Hz, 1H, ArH), 7.23-7.10 (m, 5H, ArH), 7.05-6.85 (m, 8H, ArH), 6.40 (d, *J* = 8.4 Hz, 1H, ArH), 5.38 (s, 1H, CH), 3.49 (s, 3H, $CH_3$), 3.20 (s, 3H, $CH_3$), 3.08 (s, 3H, $CH_3$), 2.24 (s, 6H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 197.9, 171.2, 165.6, 149.7, 142.2, 140.6, 138.2, 137.5, 136.7, 135.5, 134.7, 132.7, 129.4, 128.6, 128.4, 128.3, 125.6, 124.6, 122.6, 121.1, 120.6, 115.5, 109.5, 60.8, 59.5, 31.4, 28.9, 28.7, 21.5, 21.2; IR (KBr) υ: 3155, 3047, 2987, 1968, 1750, 1678, 1548，1461, 1355, 1289, 1250, 1170, 1107, 1055, 999, 935, 868, 733, 717, 590 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{39}H_{33}N_3O_4$ ([M+Na]$^+$): 630.2363, found: 630.2364.

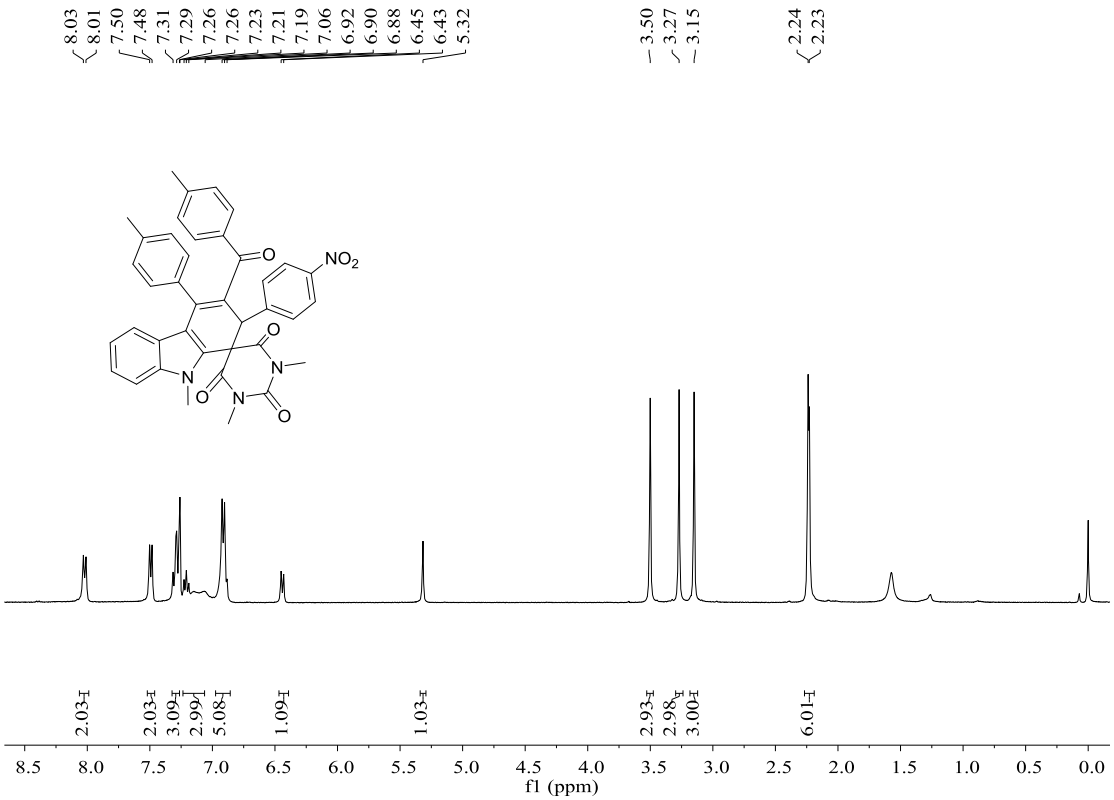


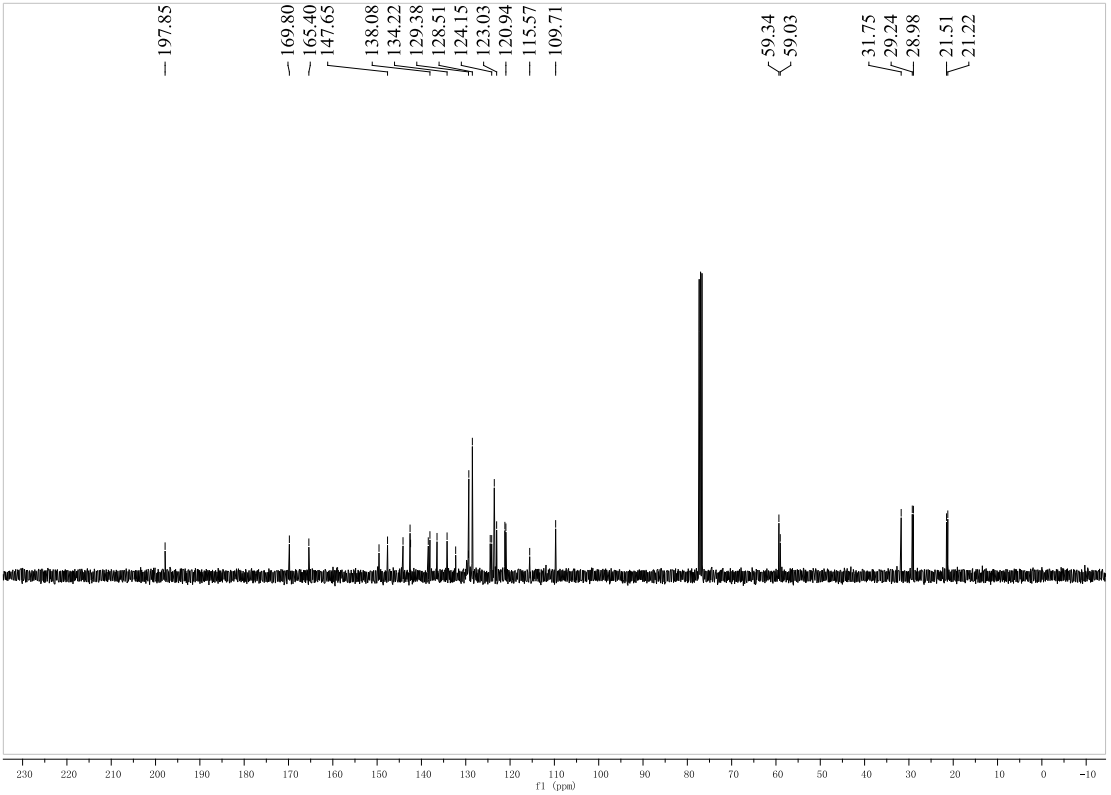

197.88
171.23
165.56
149.72
142.21
132.70
128.36
124.60
120.57
115.49
109.50
60.80
59.45
31.42
28.92
28.74
21.50
21.22
f1 (ppm)

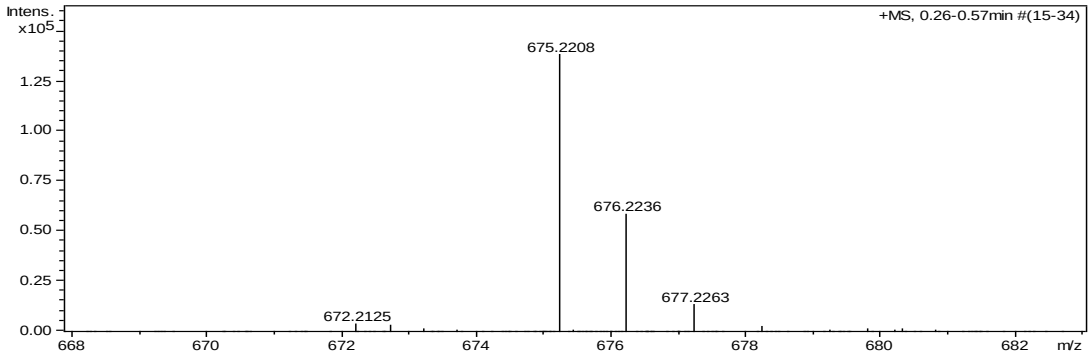

Intens.
x10^6
+MS, 0.26-0.56min #(15-33)
630.2364
631.2392
632.2415
m/z

**1',3',9-Trimethyl-3-(4-methylbenzoyl)-2-(4-nitrophenyl)-4-(*p*-tolyl)-2,9-dihydro-2'H-spiro[carbazole-1,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (5c):**

Yellow solid, 277 mg, 82%, m.p. 190-193 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 8.02 (d, *J* = 8.0 Hz, 2H, ArH), 7.49 (d, *J* = 8.0 Hz, 2H, ArH), 7.31-7.26 (m, 3H, ArH), 7.23-7.06 (m, 3H, ArH), 6.92-6.88 (m, 5H, ArH), 6.44 (d, *J* = 8.4 Hz, 1H, ArH), 5.32(s, 1H, CH), 3.50 (s, 3H, $CH_3$), 3.27 (s, 3H, $CH_3$), 3.15 (s, 3H, $CH_3$), 3.23 (d, *J* = 3.6 Hz, 6H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 197.8, 169.8, 165.4, 149.6, 147.7, 144.2, 142.6, 142.5, 142.4, 138.5, 138.1, 136.5, 134.2, 132.3, 129.4, 129.3, 129.2, 128.5, 124.5, 124.1, 123.6, 123.0, 121.2, 120.9, 115.6, 109.7, 59.3, 59.0, 31.8, 29.2, 29.0, 21.5, 21.2; IR (KBr) υ: 3156, 3041, 2966, 1932, 1780, 1625, 1566, 1438, 1369, 1286, 1233, 1165, 1173, 1049, 1062, 987, 823, 755, 713 cm$^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{39}H_{32}N_4O_6$ ($[M+Na]^+$): 675.2214, found: 675.2208.

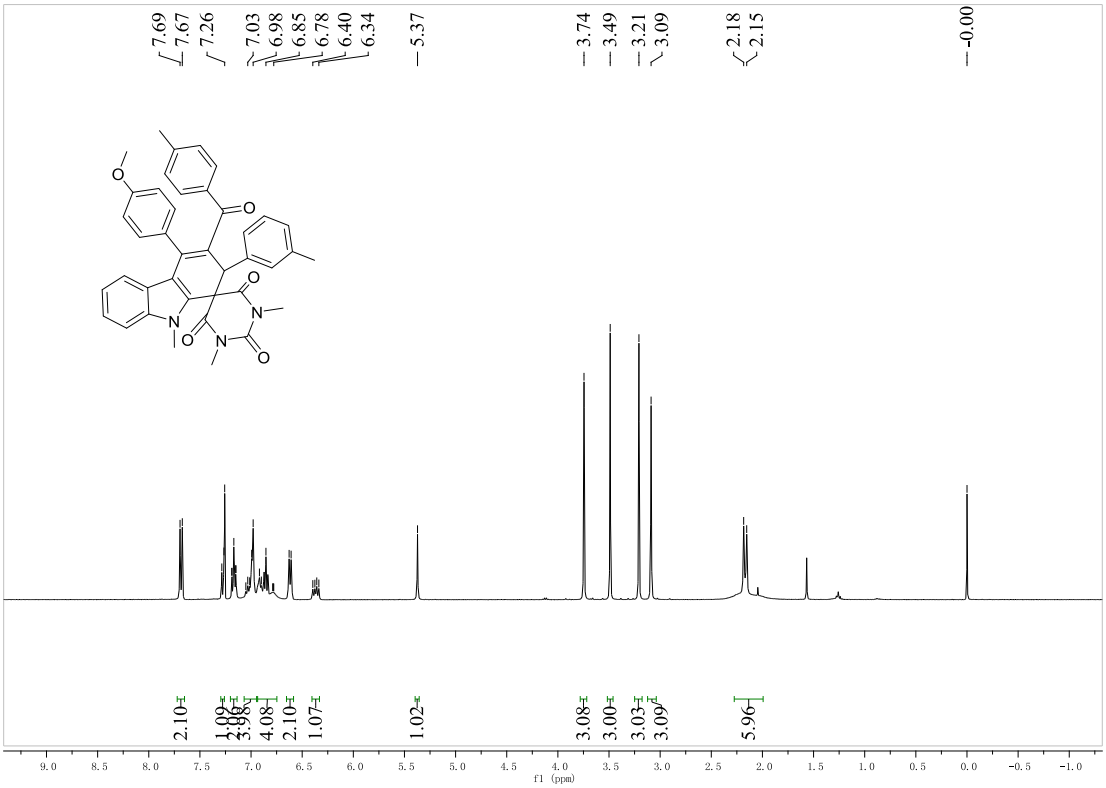


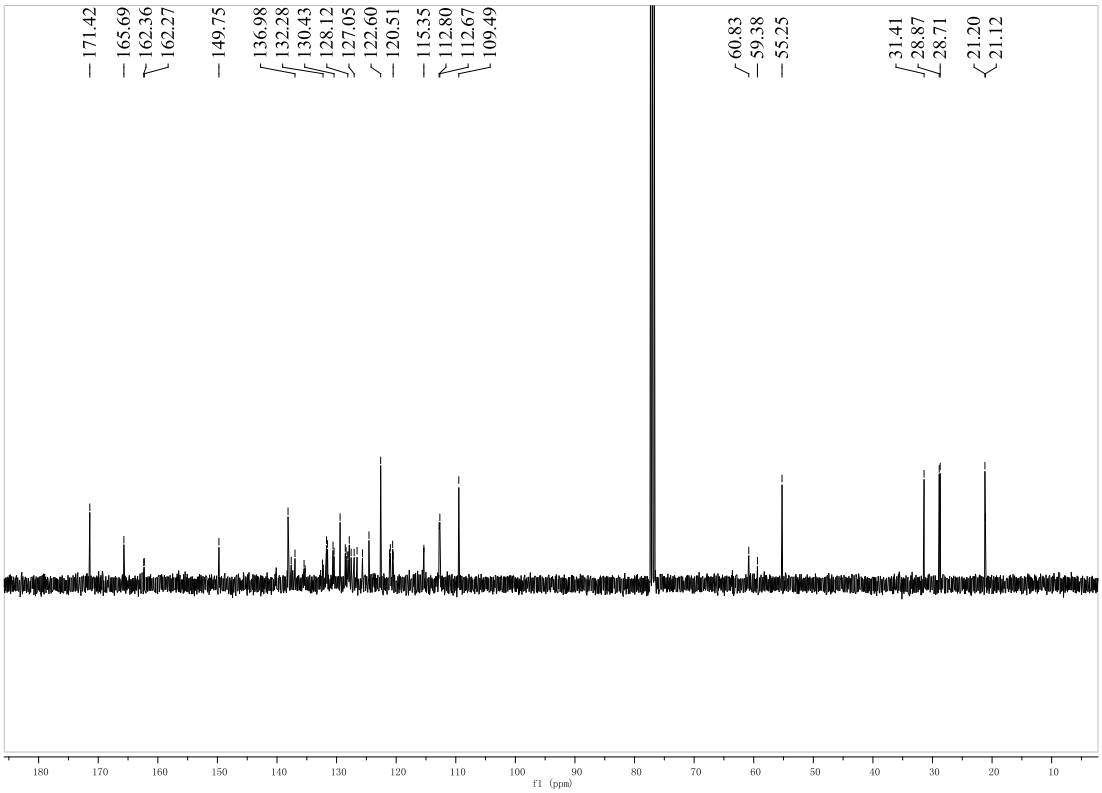

197.85
169.80
165.40
147.65
138.08
134.22
129.38
128.51
124.15
123.03
120.94
115.57
109.71
59.34
59.03
31.75
29.24
28.98
21.51
21.22
f1 (ppm)

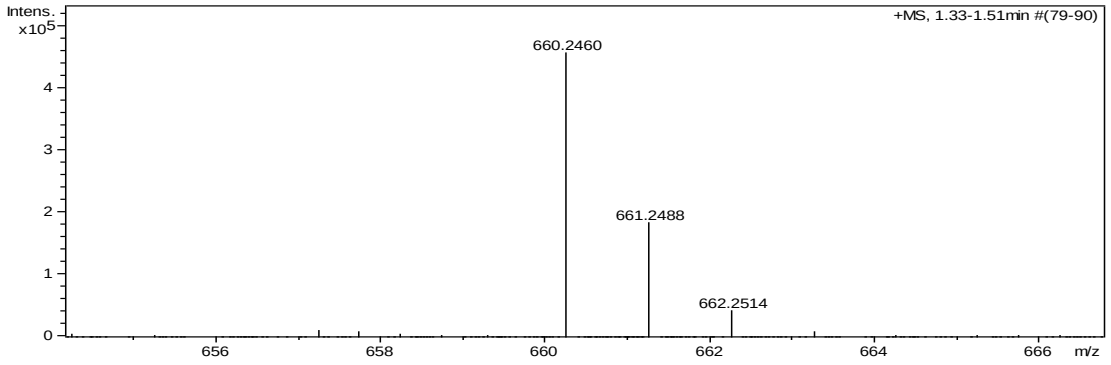

Intens.
x10^5
+MS, 0.26-0.57min #(15-34)
675.2208
676.2236
677.2263
672.2125
m/z

**4-(4-Methoxyphenyl)-1',3',9-trimethyl-3-(4-methylbenzoyl)-2-(*m*-tolyl)-2,9-dihydro-2'H-spiro[carbazole-1,5'-pyrimidine]-2',*4*',6'(1'H,3'H)-trione (5d):**

yellow solid, 267 mg, 81%, m.p. 191-194 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.68 (d, *J* = 8.8 Hz, 2H, ArH), 7.28 (d, *J* = 8.4 Hz, 1H, ArH), 7.19-7.15 (m, 2H, ArH), 7.05-6.98 (m, 4H, ArH), 6.92-6.78 (m, 4H, ArH), 6.62 (d, *J* = 7.6 Hz, 2H, ArH), 6.37 (d, *J* = 8.0 Hz, 1H, ArH), 5.37 (s, 1H, CH), 3.74 (s, 3H, $OCH_3$), 3.49 (s, 3H, $CH_3$), 3.21 (s, 3H, $CH_3$), 3.09 (s, 3H, $CH_3$), 2.17 (d, *J* = 12.0 Hz, 6H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 171.4, 165.7, 162.3, 149.7, 140.2, 138.2, 137.0, 135.4, 135.3, 132.4, 132.3, 131.5, 130.6, 130.4, 129.4, 128.5, 128.4, 127.9, 127.6, 127.1, 126.6, 125.7, 124.6, 122.6, 121.1, 121.0, 120.6, 120.5, 115.4, 112.8, 112.7, 109.5, 60.8, 59.4, 55.3, 31.4, 28.9, 28.7, 21.2, 21.1; IR (KBr) υ: 3171, 3044, 2967, 1954, 1781, 1655, 1542, 1456, 1387, 1254, 1217, 1169, 1167, 1054, 1032, 971, 888, 757, 714 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{40}H_{35}N_3O_5$ ($[M+Na]^+$): 660.2469, found: 660.2460.

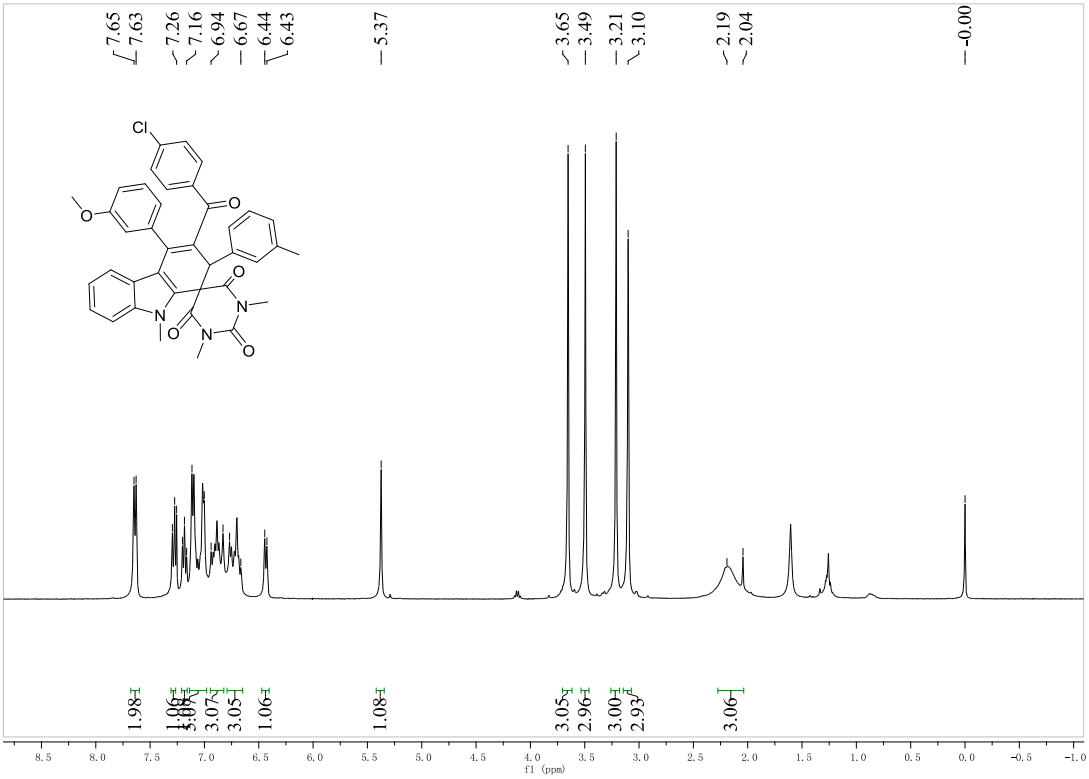


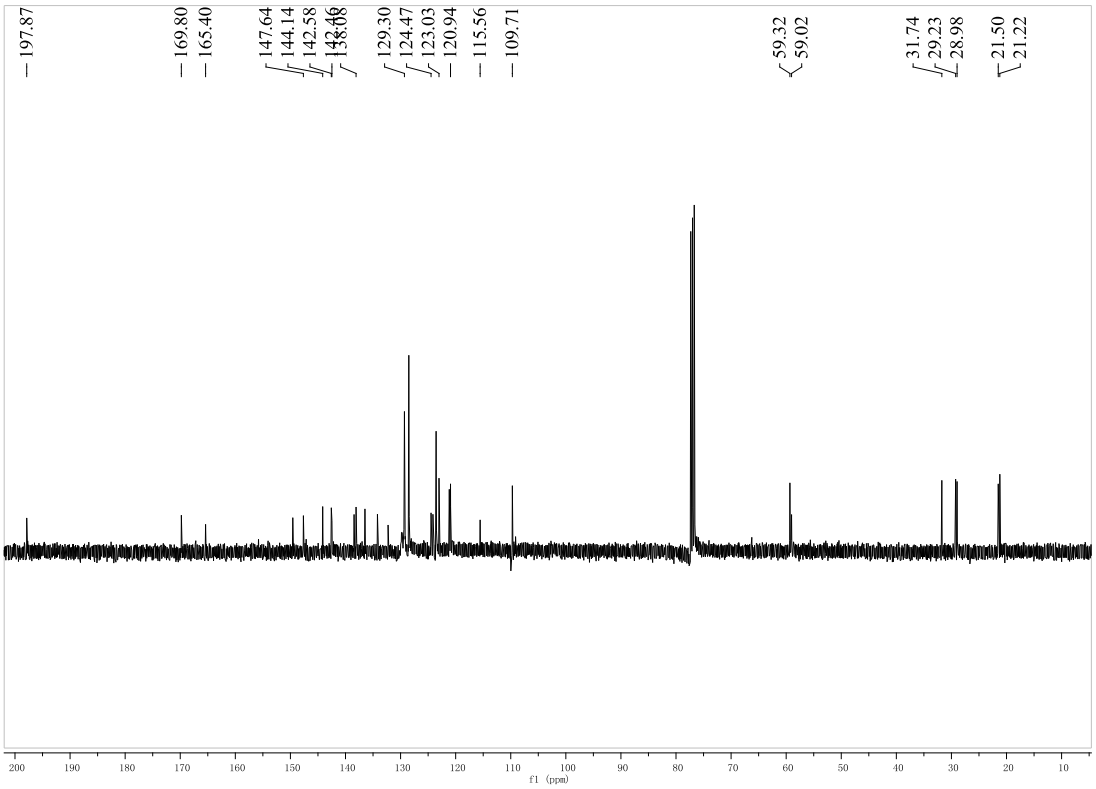

171.42
165.69
162.36
162.27
149.75
136.98
132.28
130.43
128.12
127.05
122.60
120.51
115.35
112.80
112.67
109.49
60.83
59.38
55.25
31.41
28.87
28.71
21.20
21.12
f1 (ppm)

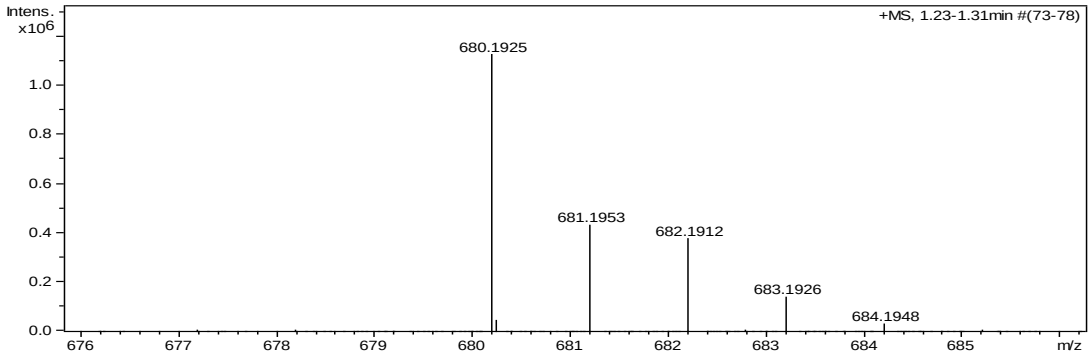

Intens.
x10^5
+MS, 1.33-1.51min #(79-90)
660.2460
661.2488
662.2514
m/z

**3-(4-Chlorobenzoyl)-4-(3-methoxyphenyl)-1',3',9-trimethyl-2-(*m*-tolyl)-2,9-dihydro-2'H-spiro [carbazole-1,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (5e):**

yellow solid, 248 mg, 73%, m.p. 187-190 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.63 (d, *J* = 7.6 Hz, 2H, ArH), 7.28 (d, *J* = 8.4 Hz, 1H, ArH), 7.18 (d, *J* = 8.0 Hz, 1H, ArH), 7.12-7.00 (m, 5H, ArH), 6.94-6.83 (m, 3H, ArH), 677.-6.67 (m, 3H, ArH), 6.43 (d, *J* = 8.0 Hz, 1H, ArH), 5.37 (s, 1H, CH), 3.65 (s, 3H, $OCH_3$), 3.49 (s, 3H, $CH_3$), 3.21 (s, 3H, $CH_3$), 3.10 (s, 3H, $CH_3$), 2.19-2.04 (m, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 197.8, 169.8, 165.3, 149.5, 147.6, 144.1, 142.5, 142.4, 138.4, 138.0, 136.4, 134.2, 132.2, 129.8, 129.7, 129.6, 129.3, 128.5, 128.4, 124.4, 124.1, 123.5, 123.0, 122.9, 121.1, 120.9, 115.5, 109.7, 59.3, 59.0, 31.7, 29.2, 28.9, 21.5, 21.2; IR (KBr) υ: 3177, 3032, 2963, 1948, 1731, 1655, 1574, 1461, 1369, 1258, 1264, 1173, 1159, 1048, 1034, 972, 867, 754, 713 $cm^{-1}$; MS (*m*/z): HRMS (ESI) Calcd. for $C_{39}H_{32}ClN_3O_5$ ($[M+Na]^+$): 680.1923, found: 680.1925.

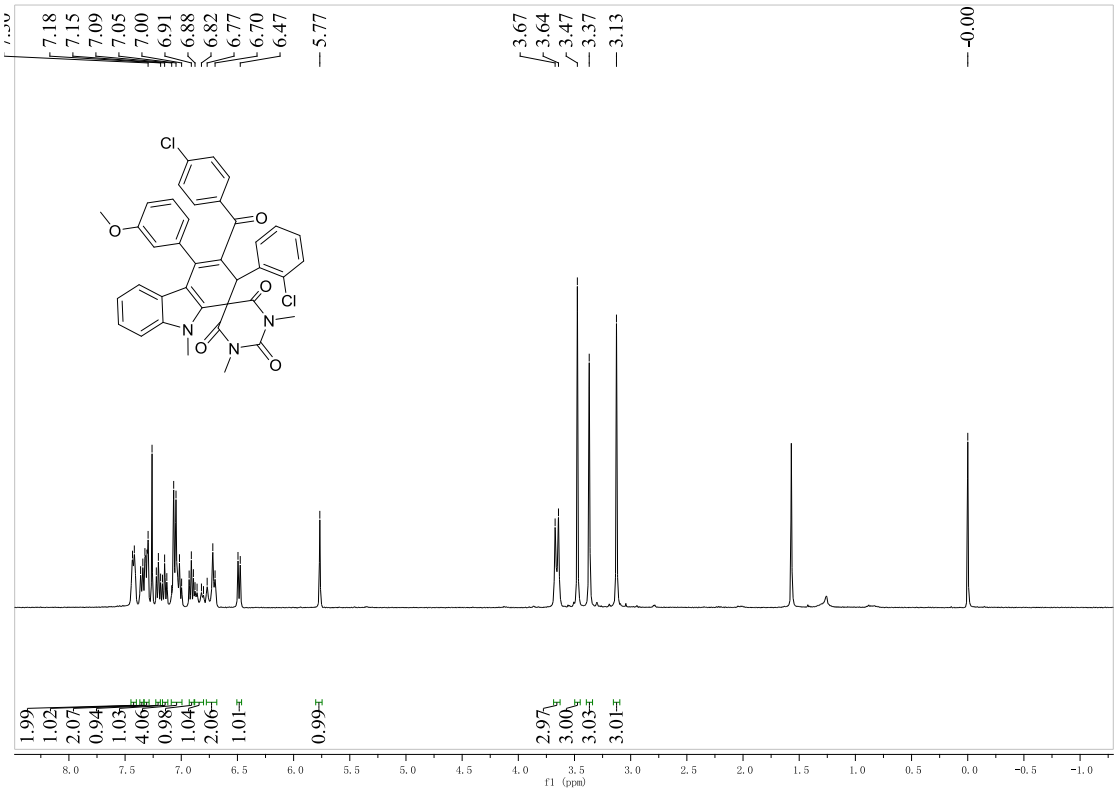


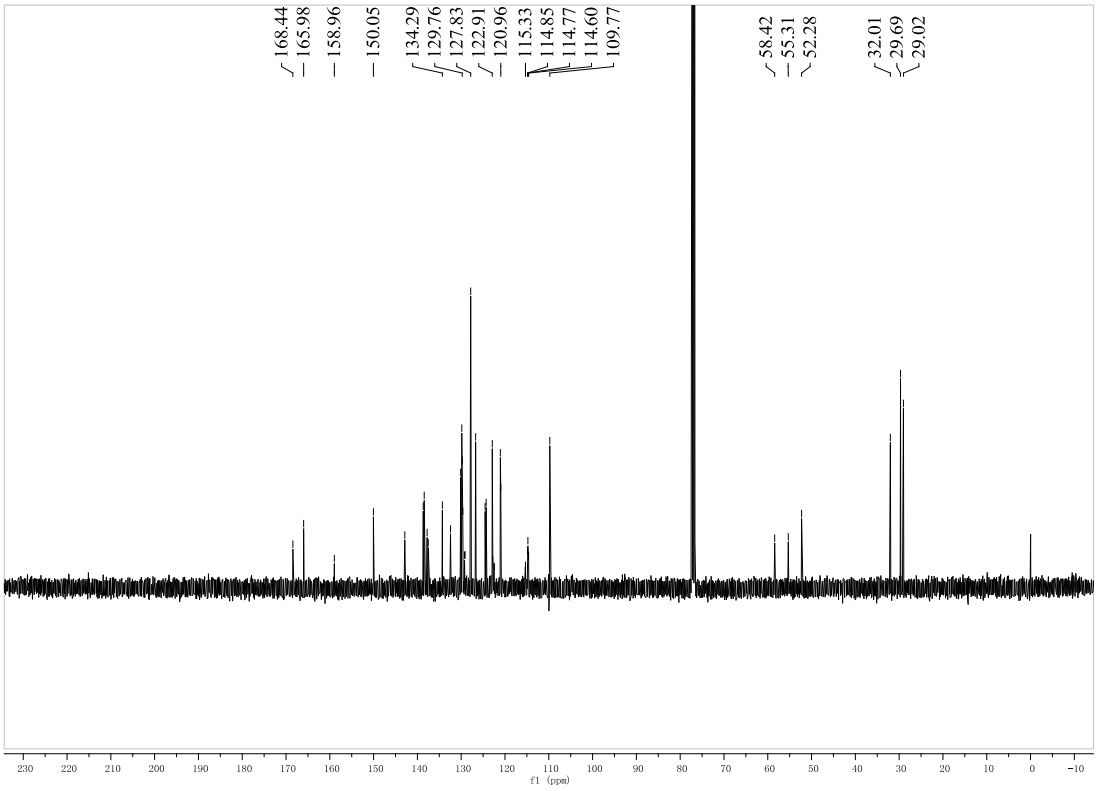

197.87
169.80
165.40
147.64
144.14
142.58
129.30
124.47
123.03
120.94
115.56
109.71
59.32
59.02
31.74
29.23
28.98
21.50
21.22
f1 (ppm)

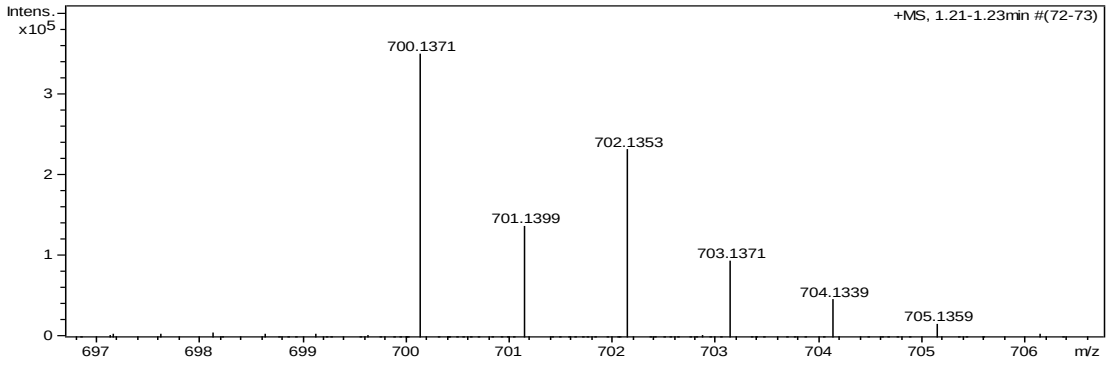

Intens.
x10^6
+MS, 1.23-1.31min #(73-78)
680.1925
681.1953
682.1912
683.1926
684.1948
m/z

**3-(4-Chlorobenzoyl)-2-(2-chlorophenyl)-4-(3-methoxyphenyl)-1',3',9-trimethyl-2,9-dihydro-2'H-spiro[carbazole-1,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (5f):**

yellow solid, 256 mg, 73%, m.p. 197-200 °C; $^1$H NMR (400 MHz, $CDCl_3$) δ: 7.43 (d, *J* = 6.4 Hz, 2H, ArH), 7.35 (d, *J* = 8.0 Hz, 1H, ArH), 7.34-7.32 (m, 2H, ArH), 7.20 (t, *J* = 7.6 Hz, 1H, ArH), 7.15 (t, *J* = 7.6 Hz, 1H, ArH), 7.09-7.00 (m, 4H, ArH), 6.91 (t, *J* = 7.6 Hz, 1H, ArH), 6.88-6.80 (m, 1H, ArH), 6.77-6.70 (m, 2H, ArH), 6.48 (d, *J* = 8.0 Hz, 1H, ArH), 5.77 (s, 1H, CH), 3.66 (d, *J* = 8.0 Hz, 3H, $OCH_3$), 3.48 (s, 3H, $CH_3$), 3.37 (s, 3H, $CH_3$), 3.13 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 168.4, 166.0, 159.0, 150.0, 142.9, 138.7, 138.4, 137.8, 137.5, 134.3, 132.4, 130.1, 129.9, 129.8, 129.6, 129.2, 129.1, 127.8, 126.7, 124.5, 124.3, 122.9, 122.8, 122.5, 121.1, 121.0, 115.3, 114.8, 114.7, 114.6, 109.8, 58.4, 55.3, 52.3, 32.0, 29.7, 29.0; IR (KBr) υ: 3164, 3055, 2973, 1967, 1745, 1693, 1556, 1431, 1356, 1254, 1263, 1149, 1168, 1071, 1056, 987, 865, 755, 765 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{38}H_{29}Cl_2N_3O_5$ ($[M+Na]^+$): 700.1376, found: 700.1371.

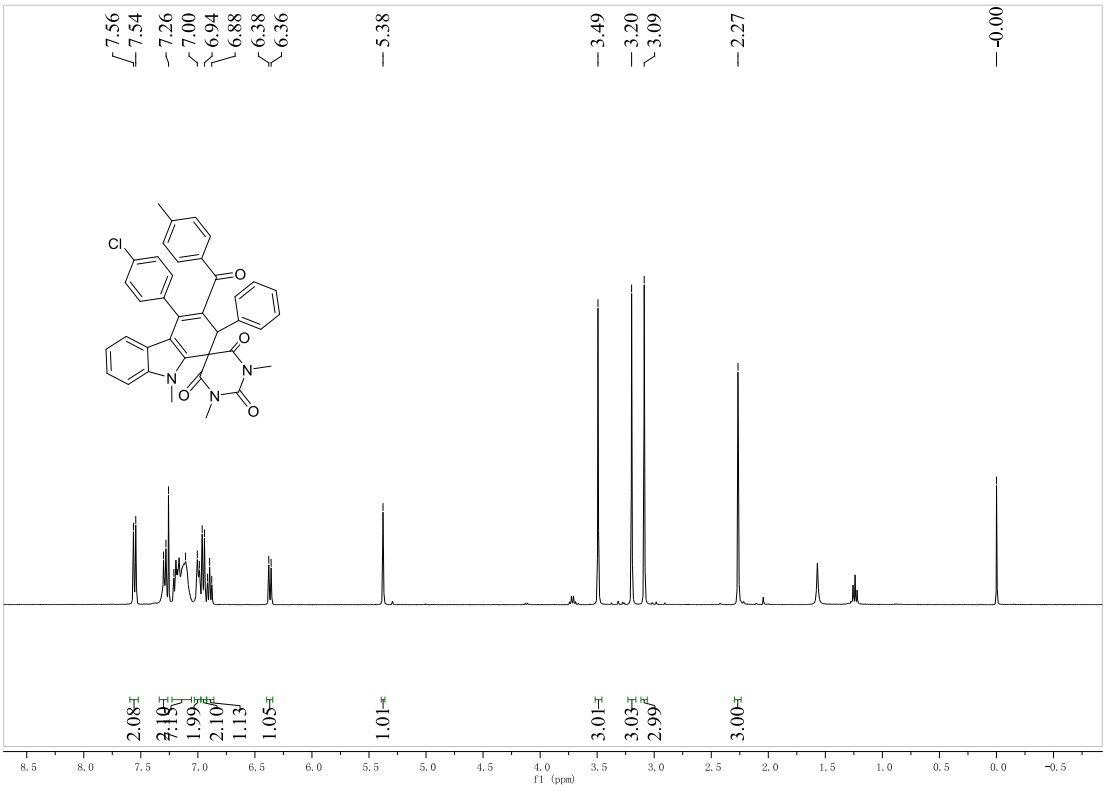


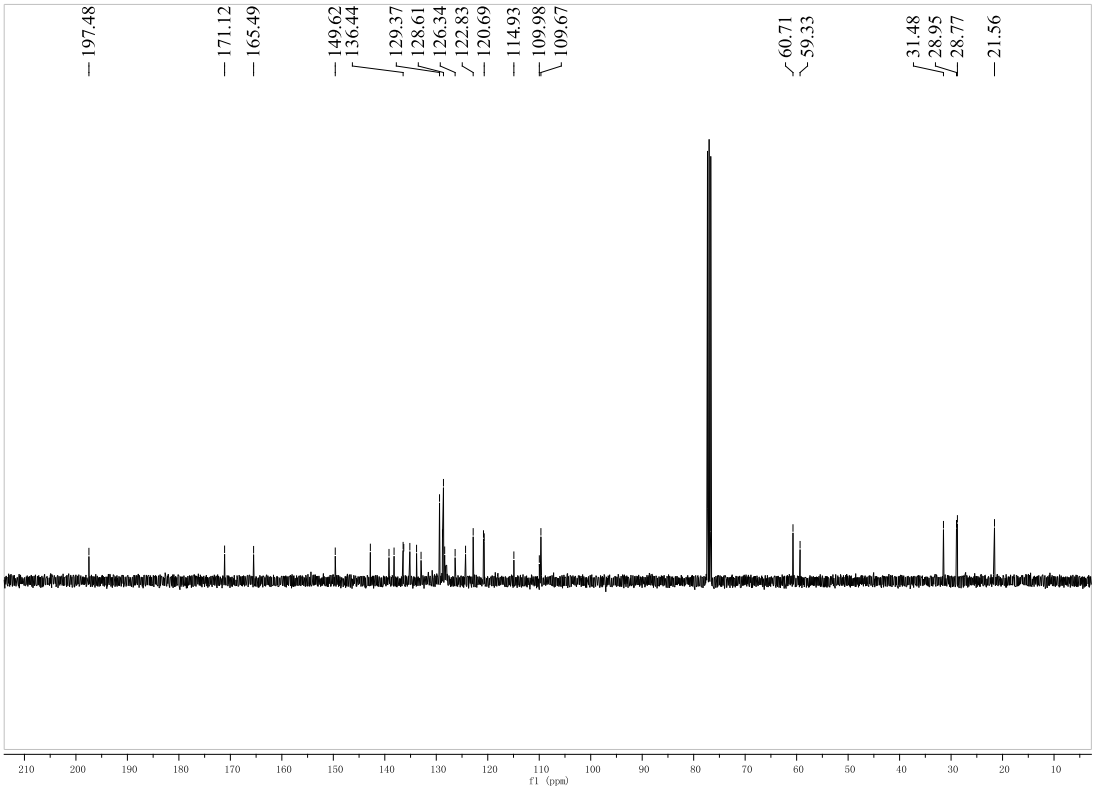

168.44
165.98
158.96
150.05
134.29
129.76
127.83
122.91
120.96
115.33
114.85
114.77
114.60
109.77
58.42
55.31
52.28
32.01
29.69
29.02
f1 (ppm)

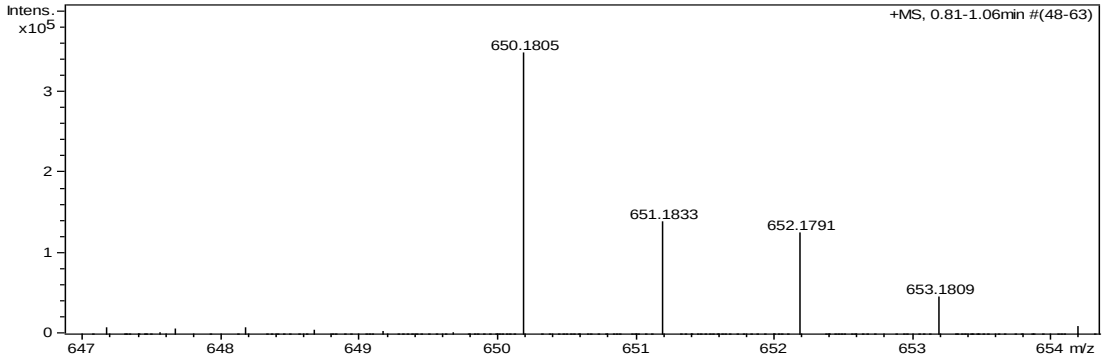

Intens.
x10^5
+MS, 1.21-1.23min #(72-73)
700.1371
702.1353
701.1399
703.1371
704.1339
705.1359
m/z

**4-(4-Chlorophenyl)-1',3',9-trimethyl-3-(4-methylbenzoyl)-2-phenyl-2,9-dihydro-2'H-spiro[carbazole-1,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (5g):**

yellow solid, 234 mg, 72%, m.p. 189-192 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.55 (d, *J* = 8.0 Hz, 2H, ArH), 7.29 (d, *J* = 8.4 Hz, 2H, ArH), 7.21-7.11 (m, 7H, ArH), 7.00 (t, *J* = 7.2 Hz, 2H, ArH), 6.95 (d, *J* = 8.0 Hz, 2H, ArH), 6.90 (d, *J* = 7.2 Hz, 2H, ArH), 6.37 (d, *J* = 8.4 Hz, 2H, ArH), 5.38 (s, 1H, CH), 3.49 (s, 3H, $CH_3$), 3.20 (s, 3H, $CH_3$), 3.09 (s, 3H, $CH_3$), 2.27 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 197.5, 171.1, 165.5, 149.6, 142.8, 139.2, 138.2, 136.4, 136.3, 135.1, 133.8, 133.0, 129.4, 128.8, 128.6, 128.4, 126.3, 124.3, 122.8, 120.8, 120.7, 114.9, 110.0, 109.7, 60.7, 59.3, 31.5, 29.0, 28.8, 21.6; IR(KBr) υ: 3144, 3082, 2973, 1965, 1748, 1642, 1578, 1467, 1356, 1245, 1214, 1171, 1145, 1069, 1055, 998, 872, 768, 749, 571 cm$^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{38}H_{30}ClN_3O_4$ ([M+Na]$^{+}$): 650.1817, found: 650.1805.

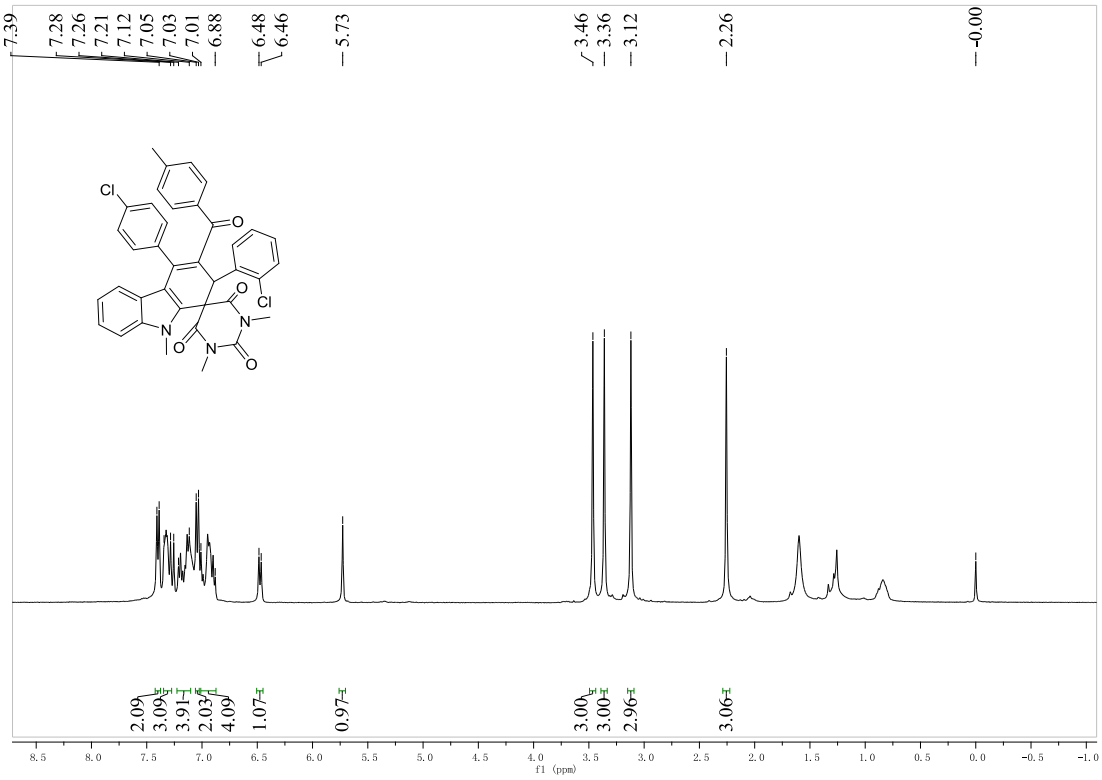


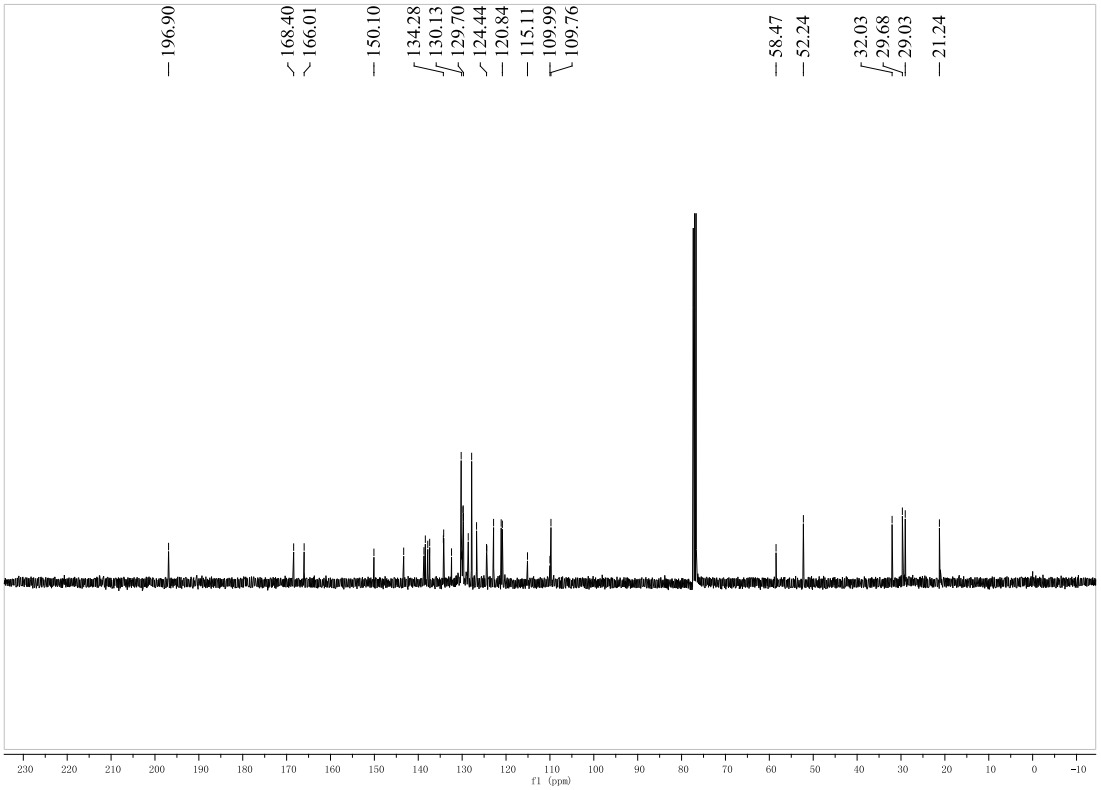

197.48
171.12
165.49
149.62
136.44
129.37
128.61
126.34
122.83
120.69
114.93
109.98
109.67
60.71
59.33
31.48
28.95
28.77
21.56
f1 (ppm)

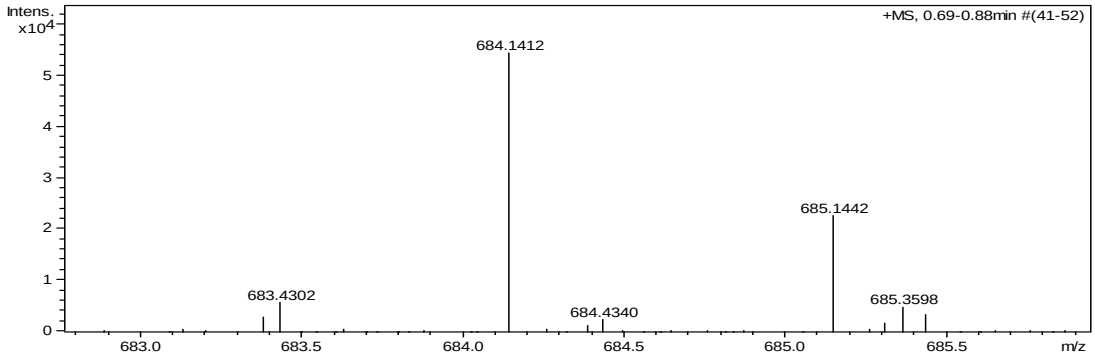

Intens.
x10^5
+MS, 0.81-1.06min #(48-63)
650.1805
651.1833
652.1791
653.1809
m/z

**2-(2-Chlorophenyl)-4-(4-chlorophenyl)-1',3',9-trimethyl-3-(4-methylbenzoyl)-2,9-dihydro-2' H-spiro[carbazole-1,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (5h):**

yellow solid, 270 mg, 79%, m.p. 190-193 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.40 (d, *J* = 8.0 Hz, 2H, ArH), 7.34-7.29 (m, 3H, ArH), 7.21-7.12 (m, 4H, ArH), 7.04 (d, *J* = 8.4 Hz, 2H, ArH), 7.01-6.88 (m, 4H, ArH), 6.48 (d, *J* = 8.0 Hz, 1H, ArH), 5.72 (s, 1H, CH), 3.46 (s, 3H, $CH_3$), 3.36 (s, 3H, $CH_3$), 3.12 (s, 3H, $CH_3$), 2.26 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 196.9, 168.4, 166.0, 150.1, 143.3, 138.7, 138.4, 137.8, 137.4, 134.3, 134.2, 132.4, 130.2, 130.1, 130.1, 129.8, 129.8, 129.7, 128.6, 127.8, 126.7, 124.4, 124.4, 122.8, 121.1, 120.8, 115.1, 110.0, 109.8, 58.5, 52.2, 32.0, 29.7, 29.0, 21.2; IR (KBr) υ: 3145, 3055, 2973, 1949, 1763, 1658, 1517, 1466, 1351, 1245, 1231, 1174, 1160, 1047, 1001, 982, 856, 741, 711 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{38}H_{30}ClN_3O_4$ ([M+Na]$^+$): 684.1427, found: 684.1412.

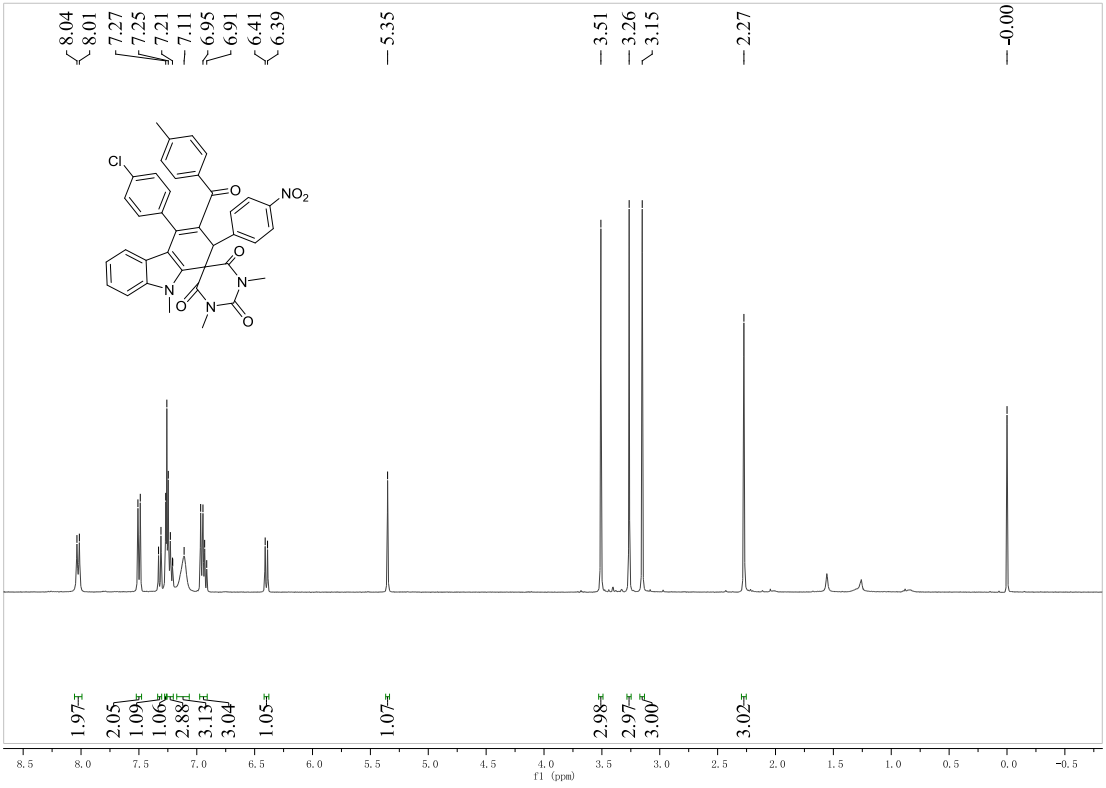


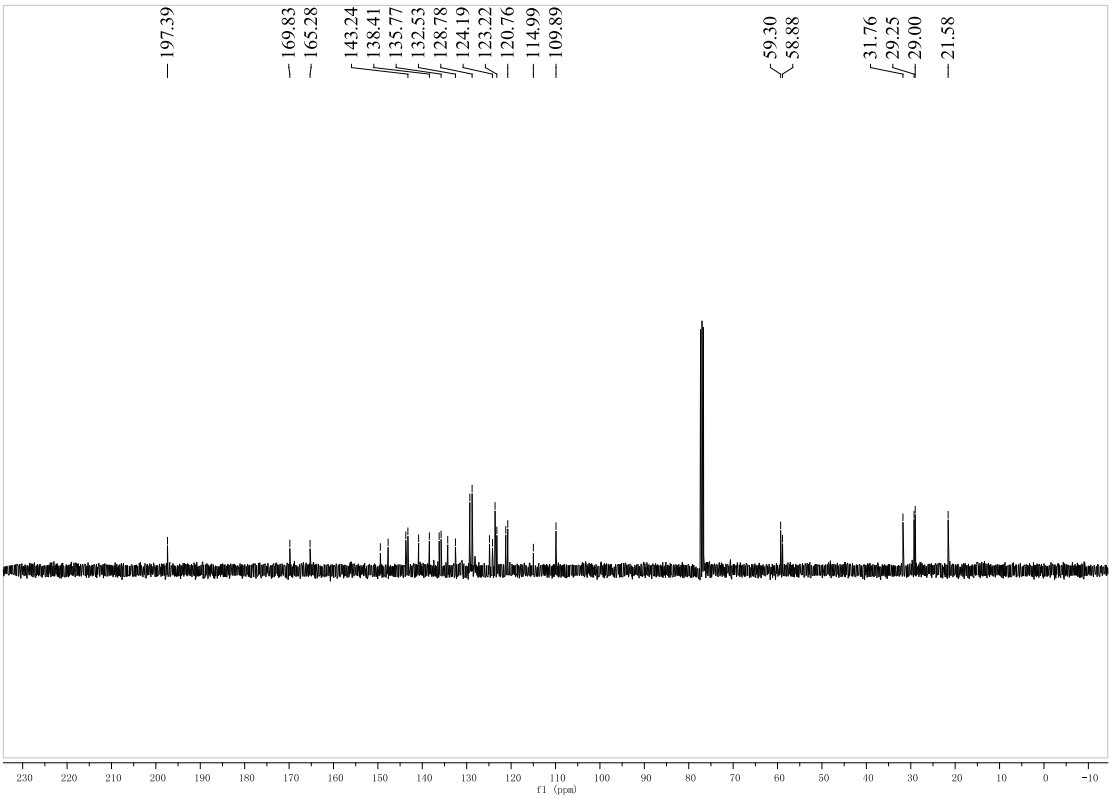

196.90
168.40
166.01
150.10
134.28
130.13
129.70
124.44
120.84
115.11
109.99
109.76
58.47
52.24
32.03
29.68
29.03
21.24
f1 (ppm)

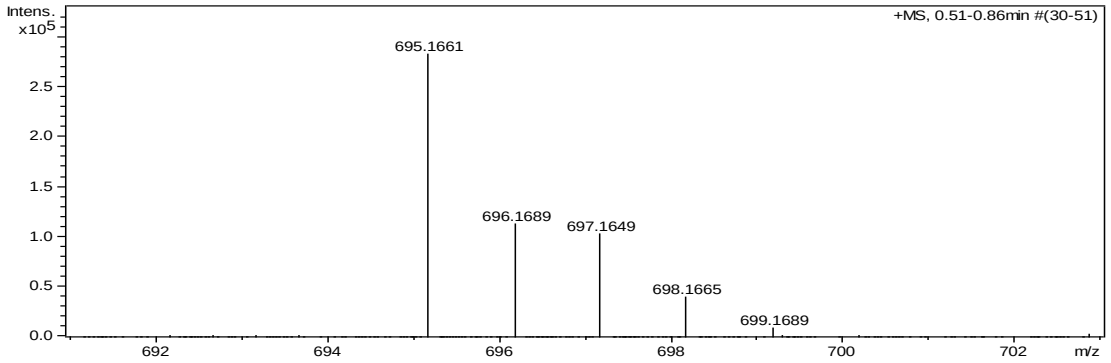

Intens.
x10^4
+MS, 0.69-0.88min #(41-52)
684.1412
685.1442
683.4302
684.4340
685.3598
m/z

**4-(4-Chlorophenyl)-1',3',9-trimethyl-3-(4-methylbenzoyl)-2-(4-nitrophenyl)-2,9-dihydro-2'H-spiro[carbazole-1,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (5i):**

yellow solid, 299 mg, 86%, m.p. 196-199 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 8.03 (d, *J* = 8.4 Hz, 2H, ArH), 7.50 (d, *J* = 8.4 Hz, 2H, ArH), 7.32 (d, *J* = 8.0 Hz, 1H, ArH), 7.27-7.21 (m,4H, ArH), 7.11 (s, 3H, ArH), 6.97-6.91 (m, 3H, ArH), 5.35 (s, 1H, CH), 3.51 (s, 3H, $CH_3$), 3.26 (s, 3H, $CH_3$), 3.15 (s, 3H, $CH_3$), 2.27 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 197.4, 169.8, 165.3, 149.4, 147.7, 143.7, 143.2, 140.8, 138.4, 136.2, 135.8, 134.3, 132.5, 129.3, 128.8, 124.9, 124.2, 123.6, 123.2, 121.2, 120.8, 115.0, 109.9, 59.3, 58.9, 31.8, 29.3, 29.0, 21.6; IR (KBr) υ: 3173, 3041, 2955, 1983, 1764, 1641, 1567, 1445, 1376, 1232, 1208, 1179, 1140, 1069, 1041, 983, 854, 766, 749 cm$^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{38}H_{30}ClN_4O_6$ ([M+Na]$^{+}$): 695.1668, found: 695.1661.

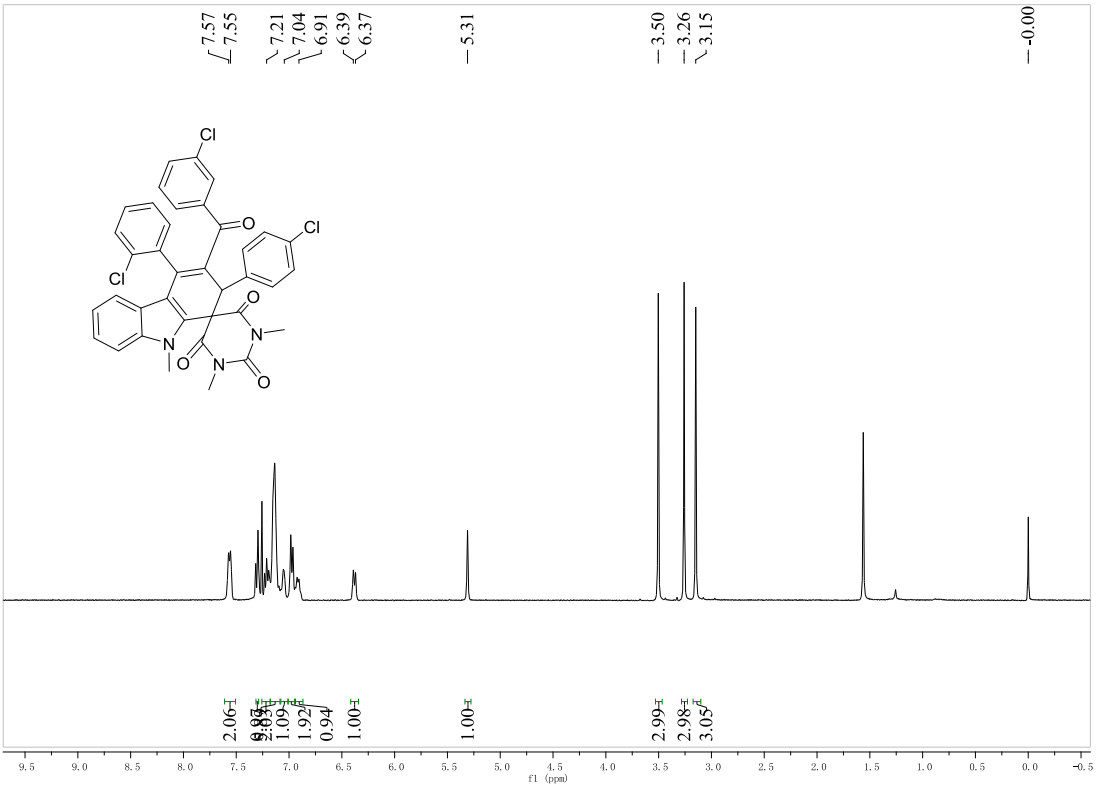


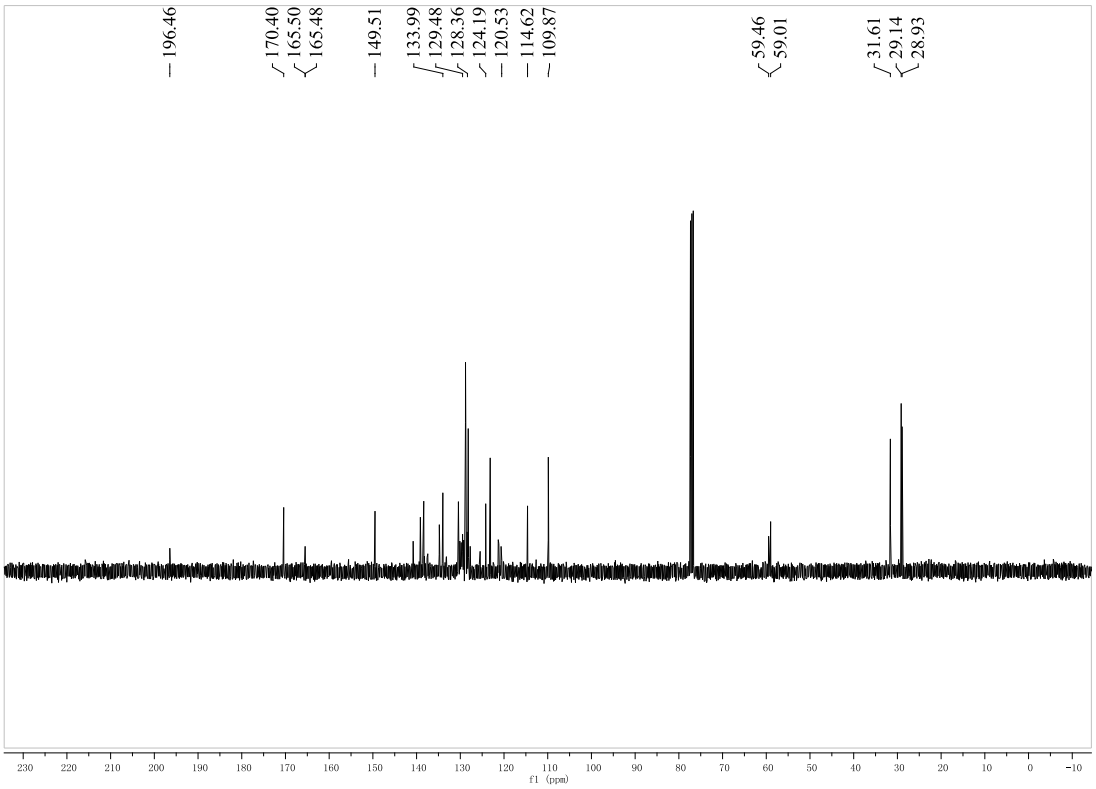

197.39
169.83
165.28
143.24
138.41
135.77
132.53
128.78
124.19
123.22
120.76
114.99
109.89
59.30
58.88
31.76
29.25
29.00
21.58
f1 (ppm)

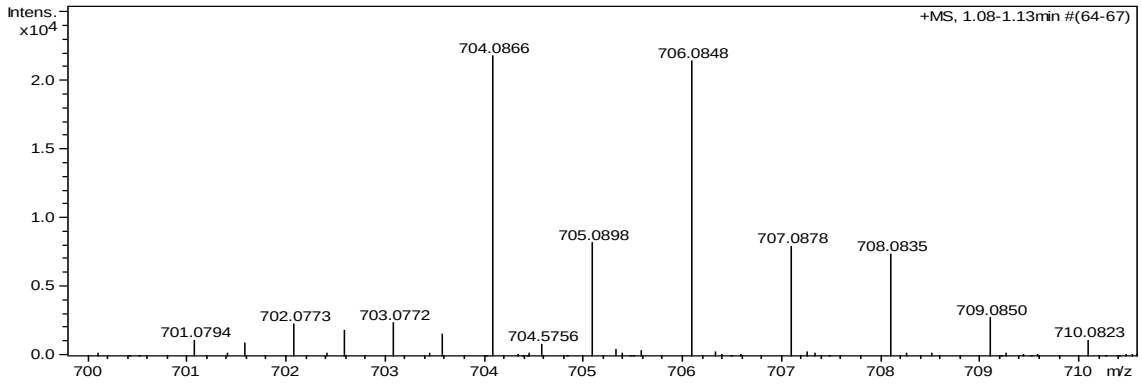

Intens.
x10^5
+MS, 0.51-0.86min #(30-51)
695.1661
696.1689
697.1649
698.1665
699.1689
m/z

**3-(3-Chlorobenzoyl)-4-(2-chlorophenyl)-2-(4-chlorophenyl)-1',3',9-trimethyl-2,9-dihydro-2'H-spiro[carbazole-1,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (5j):**

yellow solid, 264 mg, 75%, m.p. 187-189 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.56 (d, *J* = 7.2 Hz, 2H, ArH), 7.30 (d, *J* = 8.4 Hz, 1H, ArH), 7.26-7.19 (m, 2H, ArH), 7.19-7.12 (m, 6H, ArH), 7.05-7.04 (m, 1H, ArH), 6.97 (d, *J* = 8.0 Hz, 2H, ArH), 6.92-6.91 (m, 2H, ArH), 6.38 (d, *J* = 8.0 Hz, 1H, ArH), 5.31 (s, 1H, CH), 3.50 (s, 3H, $CH_3$), 3.26 (s, 3H, $CH_3$), 3.15 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 196.4, 170.4, 165.5, 165.4, 149.5, 140.7, 139.1, 138.3, 138.2, 134.8, 134.1, 133.9, 130.4, 130.1, 129.8, 129.4, 129.2, 128.8, 128.4, 128.2, 127.7, 125.5, 125.4, 124.1, 123.1, 121.3, 121.2, 120.7, 120.5, 114.6, 109.8, 59.4, 59.0, 31.6, 29.1, 28.9; IR (KBr) υ: 3153, 3049, 2976, 1983, 1792, 1663, 1571, 1455, 1382, 1293, 1271, 1146, 1131, 1045, 1033, 931, 862, 755, 738 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{37}H_{26}Cl_3N_3O_4$ ([M+Na]$^+$): 704.0881, found:704.0866.

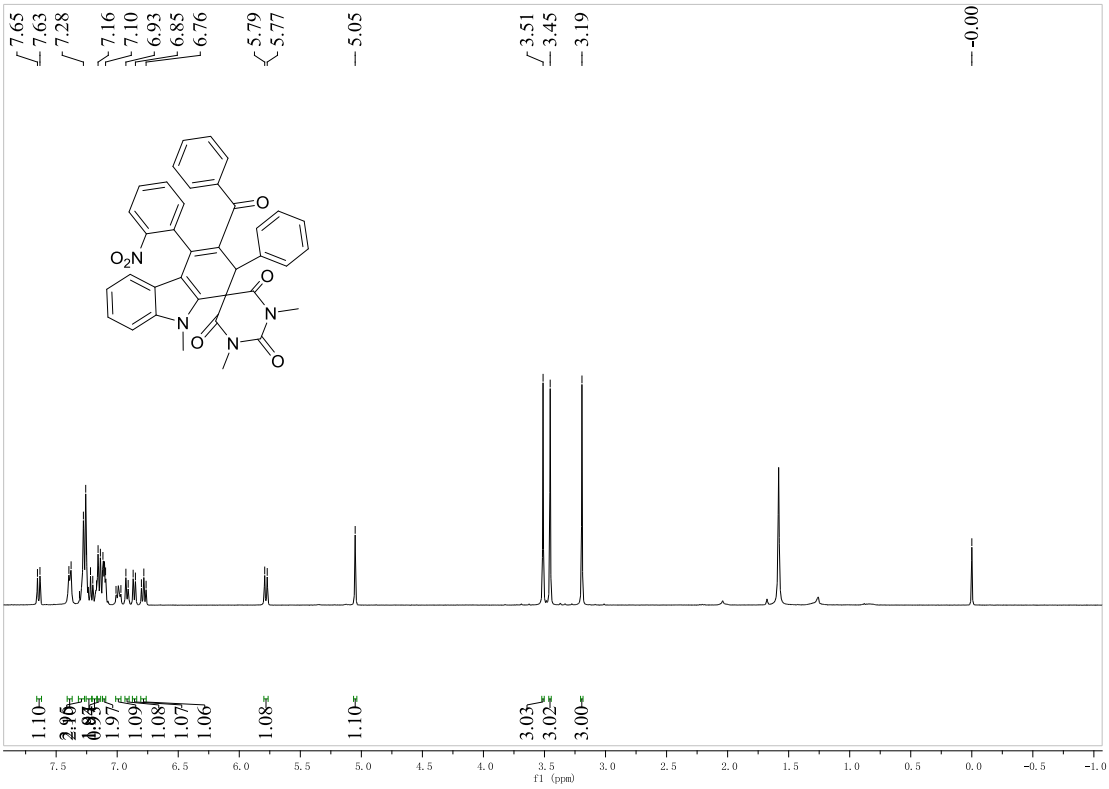


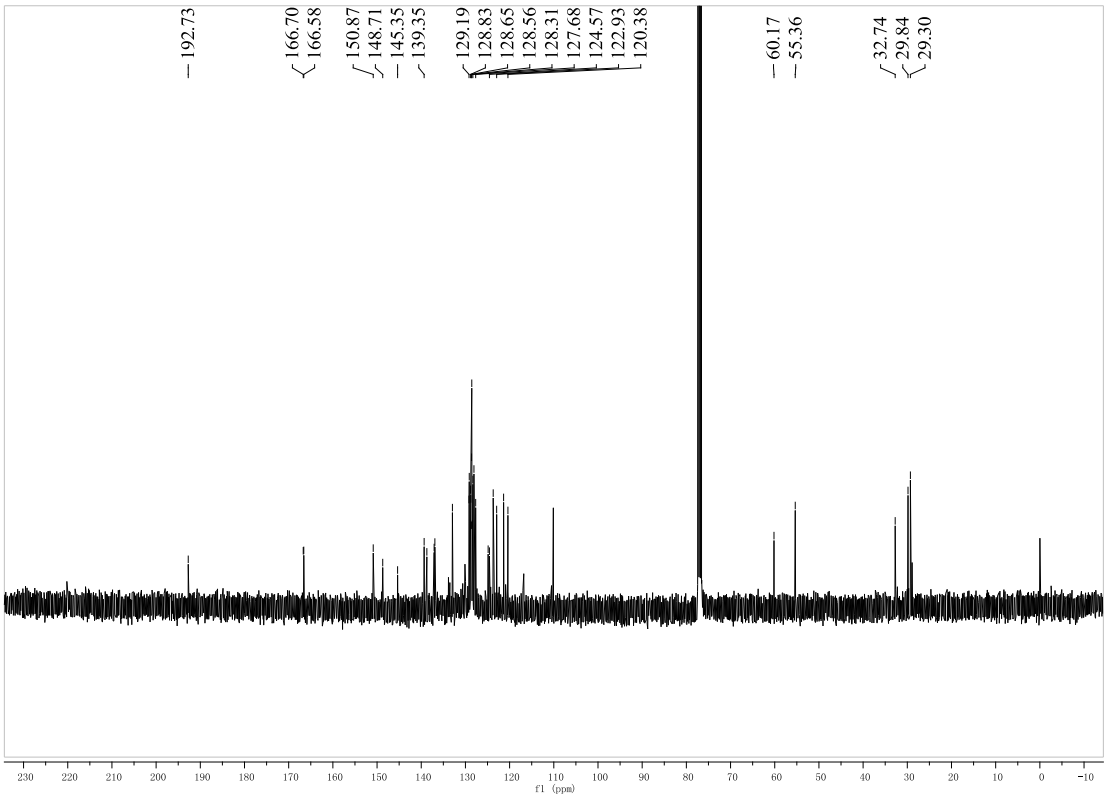

196.46
170.40
165.50
165.48
149.51
133.99
129.48
128.36
124.19
120.53
114.62
109.87
59.46
59.01
31.61
29.14
28.93
f1 (ppm)

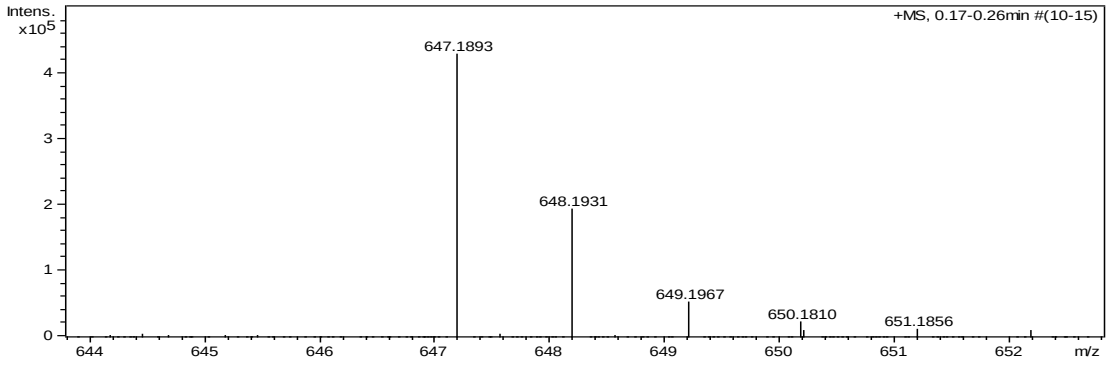

Intens.
x10^4
+MS, 1.08-1.13min #(64-67)
704.0866
706.0848
705.0898
707.0878
708.0835
709.0850
710.0823
701.0794
702.0773
703.0772
704.5756
m/z

**3-Benzoyl-1',3',9-trimethyl-4-(2-nitrophenyl)-2-phenyl-2,9-dihydro-2'H-spiro[carbazole-1,5'-pyrimidine]-2',4',6'(1'H,3'H)-trione (5k):**

yellow solid, 210 mg, 65%, m.p. 193-196 °C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: 7.64 (d, *J* = 8.0 Hz, 1H, ArH), 7.39 (d, *J* = 7.2 Hz, 2H, ArH), 7.31-7.28 (m, 3H, ArH), 7.25-7.22 (m, 2H, ArH), 7.20-7.17 (m, 1H, ArH), 7.16-7.14(m, 2H, ArH), 7.12-7.10 (m, 2H, ArH), 7.01-6.97 (m, 1H, ArH), 6.92 (d, *J* = 7.6 Hz, 1H, ArH), 6.86 (d, *J* = 7.6 Hz, 1H, ArH), 6.78 (t, *J* = 7.6 Hz, 1H, ArH), 5.78 (d, *J* = 8.4 Hz, 1H, ArH), 5.05 (s, 1H, CH), 3.51(s, 3H, $CH_3$), 3.45 (s, 3H, $CH_3$), 3.19 (s, 3H, $CH_3$); $^{13}$C NMR (100 MHz, $CDCl_3$) δ: 192.7, 166.7, 166.6, 150.9, 148.7, 145.4, 139.3, 138.7, 137.1, 136.9, 132.9, 129.2, 129.1, 128.8, 128.8, 128.7, 128.6, 128.6, 128.4, 128.3, 128.1, 127.7, 124.9, 124.6, 123.7, 122.9, 121.4, 120.4, 60.2, 55.4, 32.7, 29.8, 29.3; IR(KBr) υ: 3166, 3053, 2957, 1961, 1783, 1645, 1508, 1454, 1366, 1261, 1236, 1156, 1117, 1067, 1031, 961, 855, 745, 722 $cm^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{37}H_{28}N_4O_6$ ($[M+Na]^+$): 647.1901, found:647.1893.

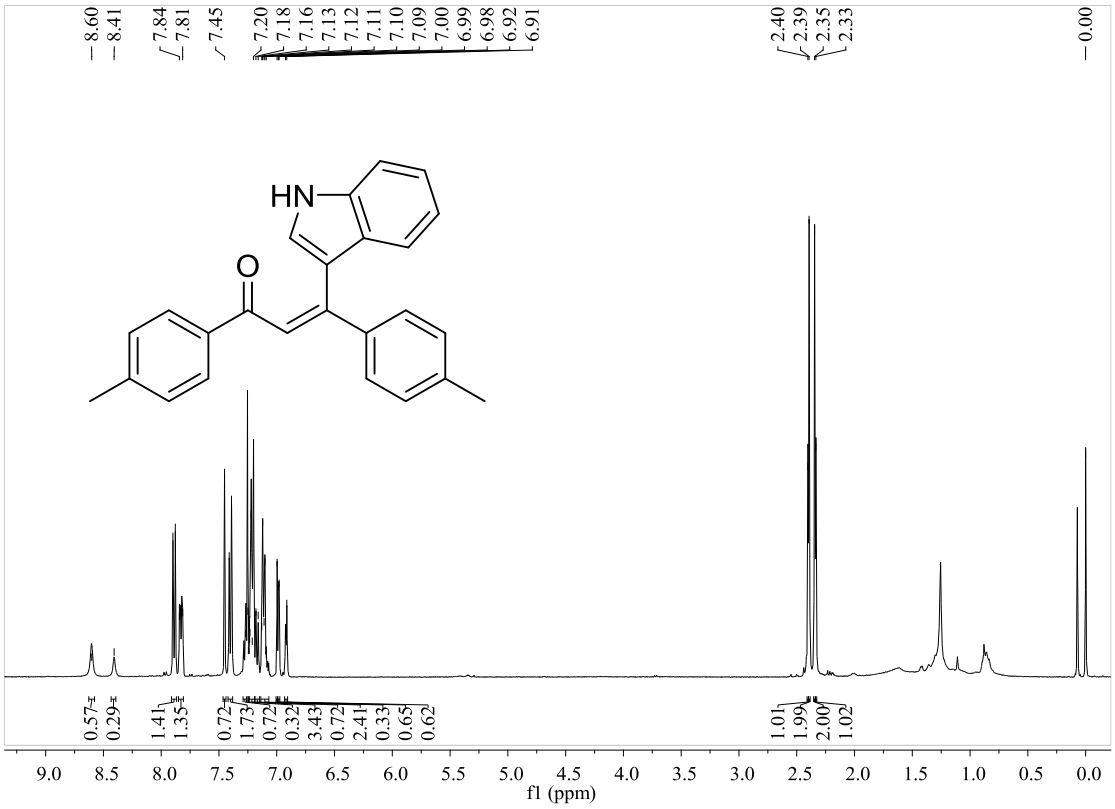


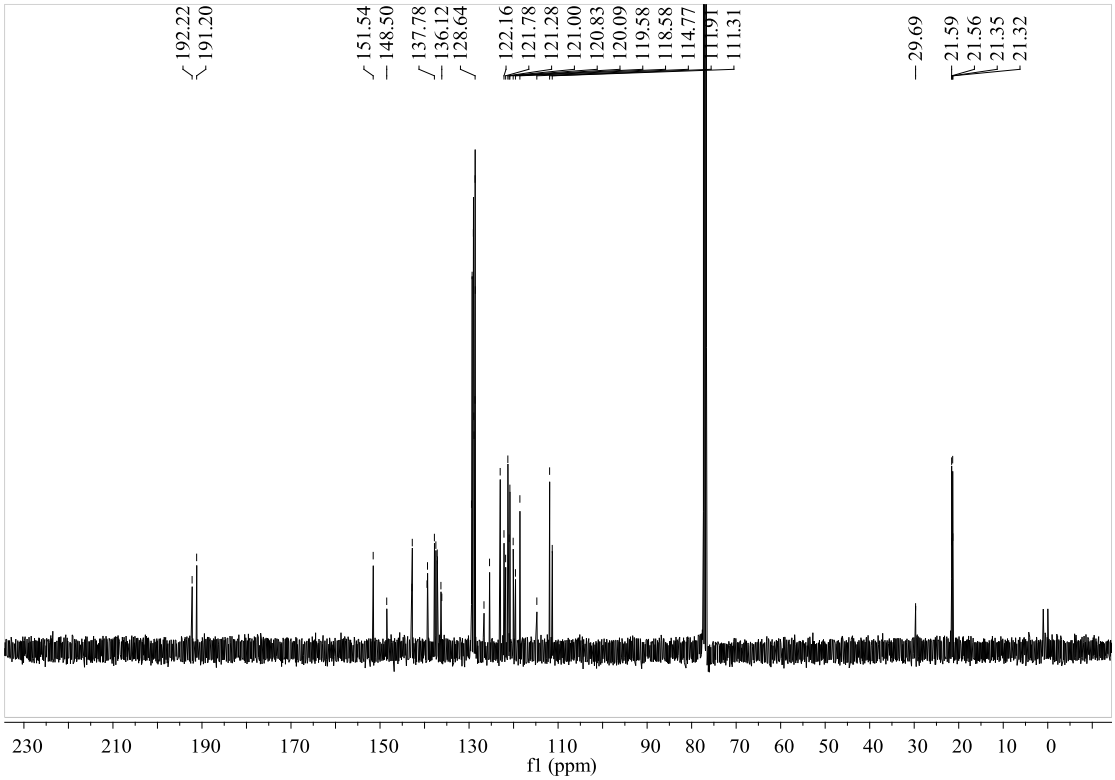

192.73
166.70
166.58
150.87
148.71
145.35
139.35
129.19
128.83
128.65
128.56
128.31
127.68
124.57
122.93
120.38
60.17
55.36
32.74
29.84
29.30
f1 (ppm)

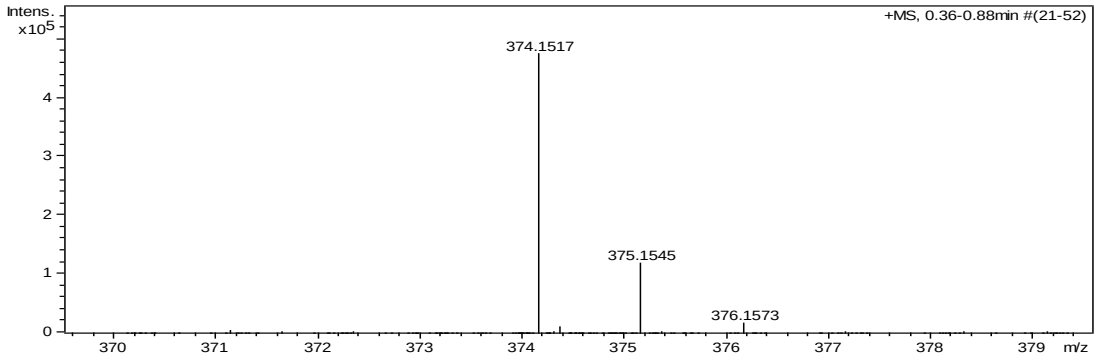

Intens.
x10^5
+MS, 0.17-0.26min #(10-15)
647.1893
648.1931
649.1967
650.1810
651.1856
m/z

**(Z)-3-(1*H*-indol-3-yl)-1,3-di-p-tolylprop-2-en-1-one (A1):**

yellow solid, 178 mg, 95%, m.p. 200-203 $^{o}$C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: **major isomer**: 8.60 (s, 1H, NH), 7.88 (d, *J* = 8.4 Hz, 1H, ArH), 7.84-7.81 (m, 2H, ArH), 7.45 (s, 1H, ArH), 7.40 (d, *J* = 8.0 Hz, 2H, ArH), 7.23-7.16 (m, 3H, ArH), 7.13-7.09 (m, 2H, ArH), 6.98 (d, *J* = 2.8 Hz, 1H, ArH), 6.91 (d, *J* = 4.4 Hz, 1H, CH), 2.39 (s, 3H, $CH_3$), 2.35 (s, 3H, $CH_3$); **minor isomer:** 8.41 (s, 1H, ArH), 7.29-7.27 (m, 2H, ArH), 7.25-7.24 (m, 1H, ArH), 7.23-7.21 (m, 1H, ArH), 7.17 (d, *J* = 7.6 Hz, 2H, ArH), 7.11-7.09 (m, 1H, ArH), 7.00 (s, 1H, ArH), 2.40 (s, 3H, $CH_3$), 2.34 (s, 3H, $CH_3$); **ratio of major/minor = 2:1**; $^{13}$C{$^{1}$H} NMR (100 MHz, $CDCl_3$) δ:192.2, 191.1, 151.5, 148.5, 142.9, 142.8, 139.5, 139.3, 137.8, 137.4, 137.2, 137.1, 136.3, 136.1, 129.4, 129.3, 129.0, 128.9, 128.8, 128.7, 128.6, 128.5, 126.7, 125.4, 123.0, 122.2, 121.8, 121.3, 121.0, 120.8, 120.0, 119.5, 118.6, 114.8, 111.9, 111.3, 29.7, 21.7, 21.6, 21.4, 21.3; IR(KBr) υ: 2913, 1905, 1732, 1611, 1540, 1417, 1350, 1217, 1206, 1145, 1132, 1017, 970, 913, 861, 747 cm$^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{25}H_{21}NO$ ([M+Na]$^{+}$): 374.1515, found: 374.1517.

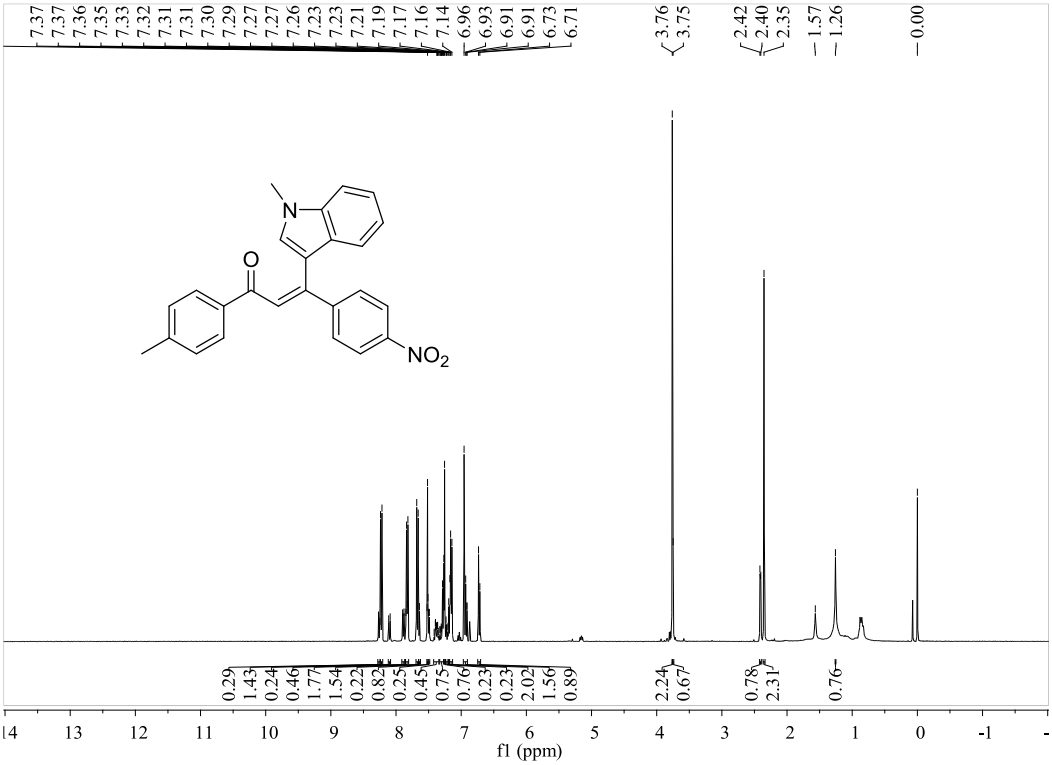


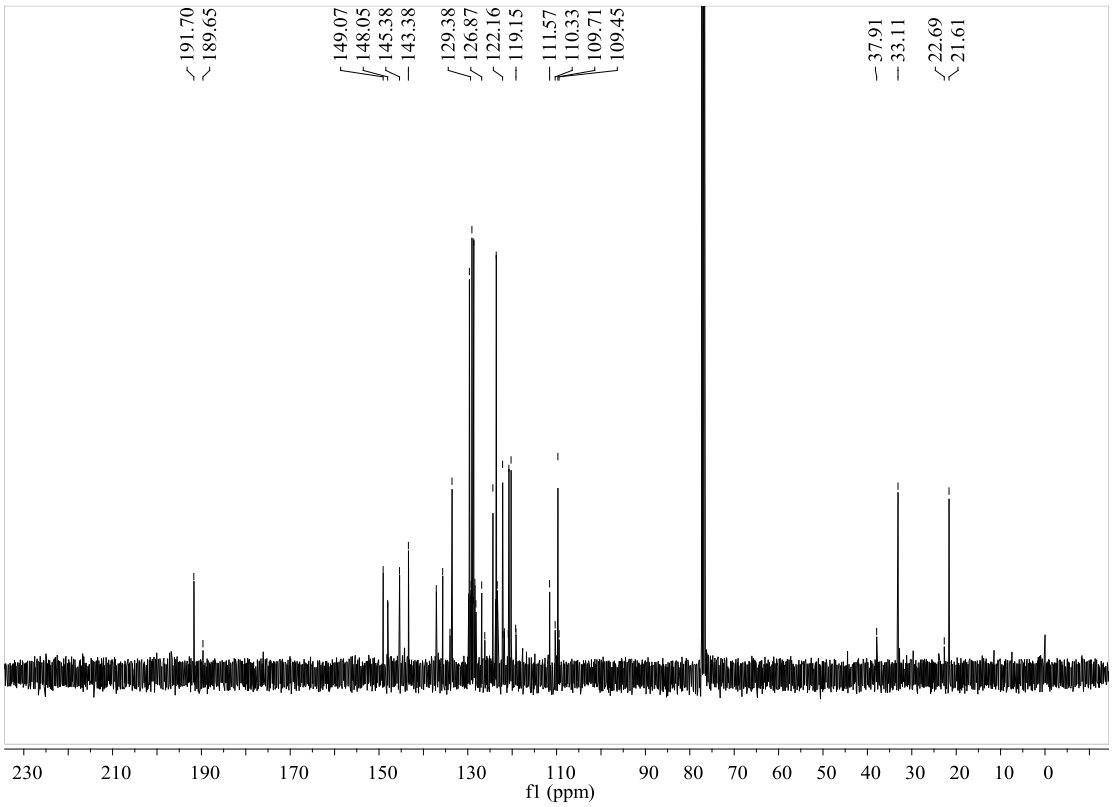

192.22
191.20
151.54
148.50
137.78
136.12
128.64
122.16
121.78
121.28
121.00
120.83
120.09
119.58
118.58
114.77
111.91
111.31
29.69
21.59
21.56
21.35
21.32
230
210
190
170
150
130
110
90
80
70
60
50
40
30
20
10
0
f1 (ppm)

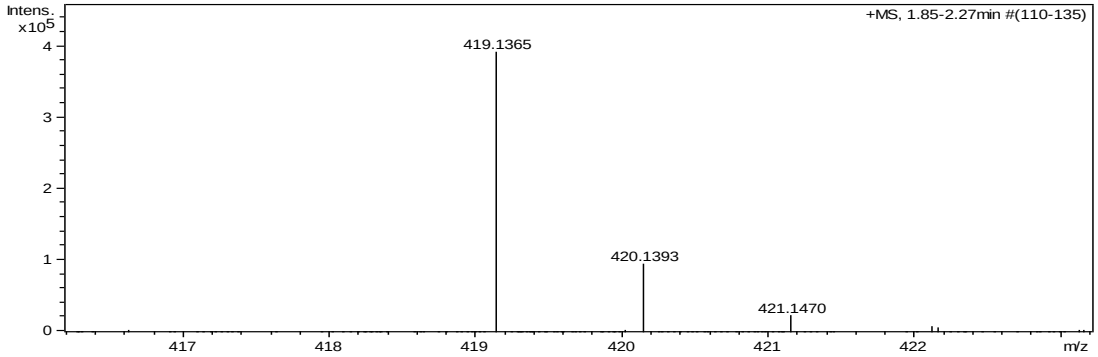

Intens.
x10^5
+MS, 0.36-0.88min #(21-52)
374.1517
375.1545
376.1573
0
1
2
3
4
370
371
372
373
374
375
376
377
378
379
m/z

**(Z)-3-(1-Methyl-1H-indol-3-yl)-3-(4-nitrophenyl)-1-(p-tolyl)prop-2-en-1-one (A2):**

yellow solid, 193 mg, 92%, m.p. 216-219 $^{o}$C; $^{1}$H NMR (400 MHz, $CDCl_3$) δ: **major isomer**: 8.22 (d, *J* = 8.8 Hz, 2H, ArH), 7.82 (d, *J* = 8.0 Hz, 2H, ArH), 7.67 (d, *J* = 8.8 Hz, 2H, ArH), 7.52 (s, 1H, ArH), 7.33-7.27 (m, 1H, ArH), 7.26 (s, 1H, ArH), 7.17-7.14 (m, 2H, ArH), 6.95-6.91 (m, 2H, ArH), 6.72 (d, *J* = 8.0 Hz, 1H, CH), 3.76 (s, 3H, $NCH_3$), 2.35 (s, 3H, $CH_3$); **minor isomer:** 8.25 (d, *J* = 8.8 Hz, 1H, ArH), 8.10 (d, *J* = 8.8 Hz, 1H, ArH), 7.88 (d, *J* = 8.4 Hz, 2H, ArH), 7.83 (d, *J* = 8.4 Hz, 1H, ArH), 7.65 (d, *J* = 7.2 Hz, 1H, ArH), 7.52-7.49 (m, 1H, ArH), 7.42-7.35 (m, 2H, ArH), 7.23-7.21 (m, 1H, ArH), 7.20 (s, 1H, CH), 3.75 (s, 3H, $NCH_3$), 2.40 (d, *J* = 5.6 Hz, 3H, $CH_3$); **ratio of major/minor = 4:1**; $^{13}$C{$^{1}$H} NMR (100 MHz, $CDCl_3$) δ: 191.7, 189.6, 149.1, 148.1, 145.4, 143.4, 137.1, 135.6, 134.0, 133.6, 129.8, 129.6, 129.3, 129.2, 129.1, 128.7, 128.6, 128.4, 128.1, 126.8, 126.2, 124.4, 123.7, 123.6, 123.4, 123.3, 122.2, 122.1, 121.8, 120.9, 120.7, 120.3, 119.2, 119.1, 111.6, 110.3, 109.7, 109.5, 37.9, 33.1, 22.7, 21.6; IR(KBr) υ: 2900, 1931, 1740, 1630, 1517, 1401, 1322, 1254, 1201, 1130, 1165, 1041, 1000, 913, 851, 747 cm$^{-1}$; MS (*m*/*z*): HRMS (ESI) Calcd. for $C_{25}H_{20}N_2O_3$ ([M+Na]$^{+}$): 419.1366, found: 419.1365.




191.70
189.65
149.07
148.05
145.38
143.38
129.38
126.87
122.16
119.15
111.57
110.33
109.71
109.45
37.91
33.11
22.69
21.61
230
210
190
170
150
130
110
90
80
70
60
50
40
30
20
10
0
f1 (ppm)


Intens.
x10^5
+MS, 1.85-2.27min #(110-135)
419.1365
420.1393
421.1470
417
418
419
420
421
422
m/z