

## Supporting information

# Diastereoselective Rhodium Catalyzed Synthesis of Indolines from *N*-Sulfonyl-1,2,3-Triazoles and *ortho*-Vinylanilines

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## **1. General Comments:**

All reactions were carried out under an atmosphere of dry nitrogen using reaction tubes. Dry toluene was prepared by distilling over Na metal and stored over using molecular sieves 4Å under N<sub>2</sub> atmosphere. All the *N*-sulfonyl-1,2,3-triazoles were synthesized from alkynes and tosyl azide employing literature procedure.<sup>1,2</sup> Similarly, all the *ortho*-vinylanilines were achieved from anilines and arylacetylenes. Rh<sub>2</sub>(OAc)<sub>4</sub>, Rh<sub>2</sub>(Oct)<sub>4</sub> and Rh<sub>2</sub>(TBSP)<sub>4</sub> were obtained from Aldrich and they were used as received.<sup>3</sup> Rh<sub>2</sub>(S-NTTL)<sub>4</sub><sup>4</sup> and Rh<sub>2</sub>(Piv)<sub>4</sub><sup>5</sup> were synthesized using literature protocol.

Column chromatography was performed using Rankem Silicagel (100-200 mesh) and the solvent system used unless otherwise specified, was ethylacetate-hexanes with various percentage of polarity depending on the nature of the substrate.

It is important to note that purity of both triazoles and dienophiles significantly affect the reactions.

## **2. Analytical Methods:**

NMR data were recorded on 400 and 500 MHz spectrometers. <sup>13</sup>C and <sup>1</sup>H NMR spectra were referenced to signals of deutero solvents and residual protonated solvents, respectively. Infrared spectra were recorded on a Thermo Nicolet iS10 FT spectrometer. HRMS were recorded by electron spray ionization (ESI) method on a Q-TOF Micro with lock spray source. Melting points are corrected. The crystal data were collected and integrated using a Bruker Axs kappa apex2 CCD diffractometer, with graphite monochromated Mo-K $\alpha$  radiation.

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<sup>1</sup> J. Raushel and V. V. Fokin, *Org. Lett.*, 2010, **12**, 4952.

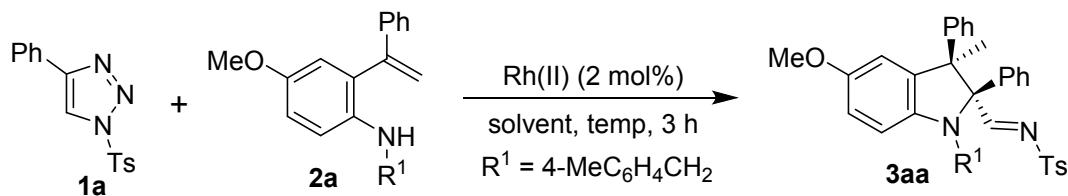
<sup>2</sup> L. Wang, S. Peng, L. J.T. Danence, Y. Gao and J. Wang, *Chem. –Eur. J.*, 2012, **18**, 6088.

<sup>3</sup> A. Arienti, F. Bigi, R. Maggi, E. Marzi, P. Moggi, M. Rastelli, G. Sartori and F. Tarantola, *Tetrahedron*, 1997, **53**, 3795.

<sup>4</sup> P. Müller, Y. Allenbach and E. Robert, *Tetrahedron Asymmetry*, 2003, **14**, 779.

<sup>5</sup> S. W. Kwok, L. Zhang, N.P. Grimster and V.V. Fokin, *Angew. Chem. Int. Ed.*, 2014, **53**, 3452.

### 3. Diastereoselective rhodium catalyzed synthesis of indolines from *N*-sulfonyl-1,2,3-triazoles and *ortho*-vinylanilines: Optimization



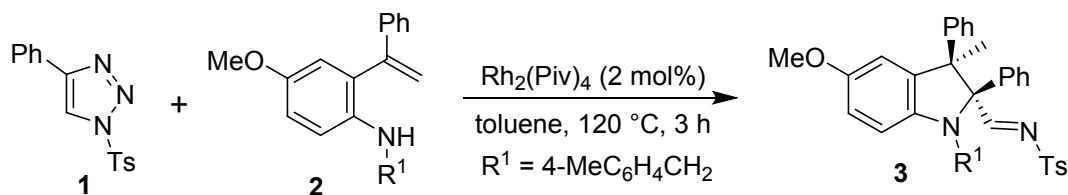
4-Phenyl-1-tosyl-1,2,3-triazole **1a** (50 mg, 0.167 mmol, 1 equiv), Rh(II)-catalyst (2 mol%) and *ortho*-vinylaniline **2a** (82 mg, 0.25 mmol, 1.5 equiv) were added under nitrogen atmosphere to an oven dried 10 mL reaction tube equipped with stir bar and solvent (1 mL) was introduced through syringe. The reaction tube was sealed and stirred at the temperature shown in the table for 3 h. After the TLC analysis, it was cooled to room temperature and the solvent was removed under reduced pressure. The resultant crude was purified by column chromatography using hexane/ethyl acetate mixture as eluent to afford indoline **3aa**.

Entry	Rh(II) (2 mol%)	Solvent	Temp (°C)	Yield (%) <sup>[a]</sup>
1	Rh <sub>2</sub> (OAc) <sub>4</sub>	Toluene	100	15
2	Rh <sub>2</sub> (Oct) <sub>4</sub>	Toluene	100	30
3	Rh <sub>2</sub> (Piv) <sub>4</sub>	Toluene	100	45
4	Rh <sub>2</sub> (S-NTTL) <sub>4</sub>	Toluene	100	5
5	Rh <sub>2</sub> (Piv) <sub>4</sub>	Benzene	100	35
6	Rh <sub>2</sub> (Piv) <sub>4</sub>	1,2-DCE	100	29
7	Rh <sub>2</sub> (Piv) <sub>4</sub>	Toluene	120	85
8	Rh <sub>2</sub> (Piv) <sub>4</sub>	Toluene	120	75 <sup>[b]</sup>

Triazole **1a** (0.167 mmol, 1 equiv), **2a** (0.25 mmol 1.5 equiv), Rh (II) (2 mol%), solvent (1 mL).

<sup>a</sup>Isolated yields. <sup>b</sup>1 mol% catalyst was used. R<sup>1</sup> = *p*-methyl benzyl

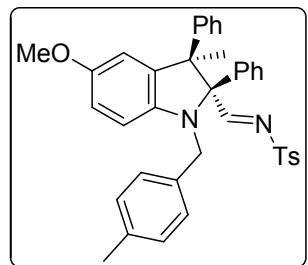
#### 4. General procedure for the diastereoselective synthesis of indolines 3:



1-Sulfonyl-1,2,3-triazole **1** (50 mg, 0.167 mmol, 1 equiv) and  $\text{Rh}_2(\text{Piv})_4$  (2 mg, 0.0033 mmol, 2 mol%) and *ortho*-vinylaniline **2** (82 mg, 0.25 mmol, 1.5 equiv) were added under nitrogen atmosphere to an oven dried 10 mL reaction tube equipped with stir bar and toluene (1 mL) was introduced through syringe. The reaction tube was sealed and stirred at 120 °C for 3 h. After the TLC analysis, it was cooled to room temperature and the solvent was removed under reduced pressure. The resultant crude was purified by column chromatography using hexane/ethyl acetate mixture as eluent to afford the indolines **3**, in diastereoselective manners.

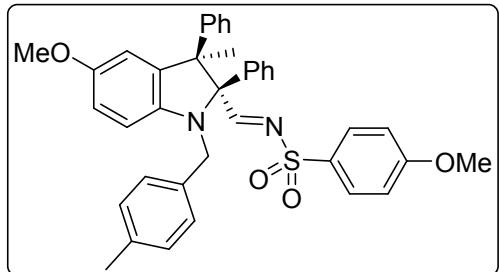
#### 5. Properties of isolated indolines 3

**Indoline (3aa):** According to general procedure the product was isolated 85% yield. Colorless gummy



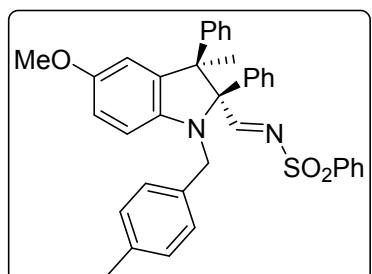
liquid;  $R_f = 0.68$  in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 3067, 2978, 2837, 1615, 1489, 1373, 1241, 1162, 1095, 916, 845, 732, 636, 549  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  8.87 (s, 1H), 7.71 (d, 2H,  $J = 8.2$  Hz), 7.28 (d, 2H,  $J = 8.0$  Hz), 7.13-7.11 (m, 4H), 7.05-6.93 (m, 8H), 6.66-6.59 (m, 3H), 6.59 (d, 1H,  $J = 2.5$  Hz), 6.20 (d, 1H,  $J = 8.4$  Hz), 3.99 (ABq, 2H,  $J = 16.6, 44.2$  Hz), 3.72 (s, 3H), 2.44 (s, 3H), 2.35 (s, 3H), 1.82 (s, 3H);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  178.5, 154.5, 144.8, 144.6, 142.3, 136.3, 136.0, 135.5, 135.1, 134.3, 129.8, 129.3, 128.2, 128.0, 127.9, 127.6, 127.6(0), 127.3, 126.5, 126.1, 113.3, 111.3, 109.4, 85.6, 57.8, 55.9, 50.0, 21.7, 21.2, 21.0; HRMS: calcd. for  $\text{C}_{38}\text{H}_{36}\text{N}_2\text{O}_3\text{S}+\text{H}$ : 601.2519; found: 617.2510.

**Indoline (3ba):** According to general procedure the product was isolated 83% yield. White solid;  $R_f$  =



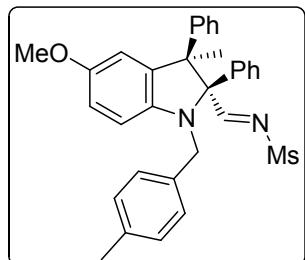
0.52 in 20% ethyl acetate/hexane; mp: 171-173 °C; FTIR (KBr): 3058, 2970, 2935, 2836, 1597, 1490, 1374, 1327, 1263, 1156, 1092, 1031, 809, 762, 735, 556, 487 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C):  $\delta$  8.85 (s, 1H), 7.75 (d, 2H,  $J$  = 8.9 Hz), 7.14-7.12 (m, 4H), 7.05-6.92 (m, 10H), 6.65-6.62 (m, 3H), 6.58 (d, 1H,  $J$  = 2.5 Hz), 6.20 (d, 1H,  $J$  = 8.5 Hz), 3.99 (ABq, 2H,  $J$  = 16.6, 42.7 Hz), 3.88 (s, 3H), 3.72 (s, 3H), 2.35 (s, 3H), 1.81 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C):  $\delta$  177.8, 163.8, 154.5, 144.7, 142.4, 136.3, 136.0, 135.2, 134.5, 130.2, 129.7, 129.3, 128.2, 127.9, 127.6, 127.5, 127.3, 126.5, 126.2, 114.5, 113.2, 111.3, 109.3, 85.5, 57.9, 55.9, 55.7, 50.1, 21.2, 21.0; HRMS: calcd. for C<sub>38</sub>H<sub>36</sub>N<sub>2</sub>O<sub>4</sub>S+H: 617.2469; found: 617.2453.

**Indoline (3ca):** According to general procedure the product was isolated 77% yield. White solid;  $R_f$  =



0.68 in 20% ethyl acetate/hexane; mp: 155-157 °C; FTIR (KBr): 3059, 2925, 2833, 1615, 1489, 1379, 1267, 1199, 1089, 1036, 969, 902, 806, 772, 699, 608, 576, 523, 421 cm<sup>-1</sup>. <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C):  $\delta$  8.92 (s, 1H), 7.82 (d, 2H,  $J$  = 8.5 Hz), 7.64-7.59 (m, 1H), 7.50-7.46 (m, 2H), 7.12-7.10 (m, 4H), 7.05-6.93 (m, 8H), 6.65-6.59 (m, 4H), 6.20 (d, 1H,  $J$  = 8.4 Hz), 3.99 (ABq, 2H,  $J$  = 16.5, 41.4 Hz), 3.72 (s, 3H), 2.35 (s, 3H), 1.82 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C):  $\delta$  179.1, 154.6, 144.6, 142.3, 138.5, 136.3, 136.0, 135.1, 134.4, 133.7, 129.4, 129.2, 128.2, 128.0, 127.9, 127.7, 127.6, 127.4, 126.5, 126.2, 113.3, 111.3, 109.4, 85.7, 58.0, 56.0, 50.1, 21.2, 21.0; HRMS: calcd. for C<sub>37</sub>H<sub>34</sub>N<sub>2</sub>O<sub>3</sub>S+H: 587.2368; found: 587.2363.

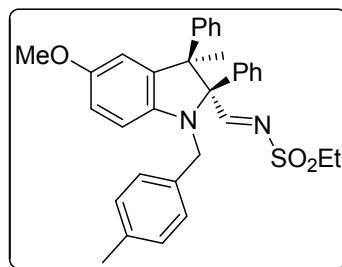
**Indoline (3da):** According to general procedure the product was isolated 90% yield. White solid;  $R_f$  =



0.56 in 20% ethyl acetate/hexane; mp: 178-180 °C; FTIR (KBr): 3056, 2983, 2930, 1617, 1490, 1428, 1322, 1265, 1152, 1036, 967, 897, 740, 516, 408 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C):  $\delta$  8.87 (s, 1H), 7.24 (d, 2H,  $J$  = 8.9

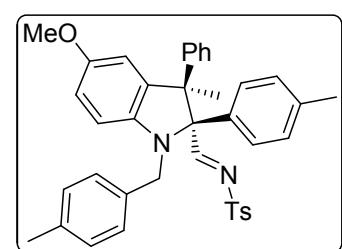
Hz), 7.16 (d, 2H,  $J$  = 7.9 Hz), 7.10-6.94 (m, 8H), 6.68-6.63 (m, 4H), 6.26 (d, 1H,  $J$  = 8.3 Hz), 4.18 (ABq, 2H,  $J$  = 9.1, 16.6 Hz), 3.73 (s, 3H), 2.99 (s, 3H), 2.36 (s, 3H), 1.91 (s, 3H);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  179.4, 154.7, 144.4, 142.3, 136.4, 136.0, 135.1, 134.3, 129.4, 128.2, 128.0, 127.7, 127.7(4), 127.4, 126.5, 126.2, 113.4, 111.4, 109.6, 85.5, 57.8, 56.0, 50.0, 40.6, 21.2, 21.0; HRMS: calcd. for  $\text{C}_{32}\text{H}_{32}\text{N}_2\text{O}_3\text{S}+\text{Na}$ : 547.2026; found: 547.2051.

**Indoline (3ea):** According to general procedure the product was isolated 88% yield. White solid;  $R_f$  =



0.56 in 20% in ethyl acetate/hexane; mp: 176-178 °C; FTIR (KBr): 3058, 2973, 2929, 2837, 1620, 1489, 1449, 1322, 1275, 1145, 1038, 970, 793, 742, 704, 544  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  8.95 (s, 1H), 7.24 (d, 2H,  $J$  = 8.5 Hz), 7.15 (d, 2H,  $J$  = 7.9 Hz), 7.10-6.95 (m, 8H), 6.67-6.63 (m, 4H), 6.26 (d, 1H,  $J$  = 8.4 Hz), 4.19 (ABq, 2H,  $J$  = 16.6, 21.9 Hz), 3.72 (s, 3H), 3.13-3.03 (m, 2H), 2.35 (s, 3H), 1.91 (s, 3H), 1.26 (t, 3H,  $J$  = 7.4 Hz);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  179.9, 154.6, 144.7, 142.4, 136.4, 136.0, 135.3, 134.6, 129.4, 128.2, 127.9, 127.8, 127.7, 127.4, 126.5, 126.2, 113.4, 111.3, 109.5, 85.7, 58.0, 56.0, 50.2, 47.2, 21.2, 21.1, 7.7; HRMS: calcd. for  $\text{C}_{33}\text{H}_{34}\text{N}_2\text{O}_3\text{S}+\text{H}$ : 539.2363; found: 539.2374.

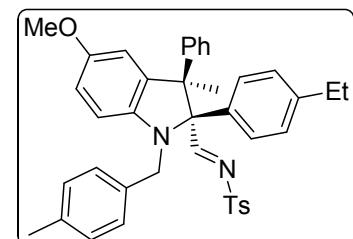
**Indoline (3fa):** According to general procedure the product was isolated 72% yield. Colorless liquid;



$R_f$  = 0.68 in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 3058, 2972, 2931, 2834, 1613, 1489, 1442, 1330, 1284, 1164, 1126, 1090, 1038, 807, 741, 714, 549, 484  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  8.86 (s, 1H), 7.68 (d, 2H,  $J$  = 8.2 Hz), 7.26 (d, 2H,  $J$  = 8.6 Hz), 7.11-7.08 (m, 4H), 6.99-6.95 (m, 3H), 6.85-6.74 (m, 4H), 6.63-6.56 (m, 4H), 6.17 (d, 1H,  $J$  = 8.4 Hz), 3.97 (ABq, 2H,  $J$  = 16.3, 25.6 Hz), 3.71 (s, 3H), 2.43 (s, 3H), 2.33 (s, 3H), 2.15 (s, 3H), 1.78 (s, 3H);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  178.7, 154.4, 144.7, 144.7(3), 142.4, 137.3, 136.2, 135.9, 135.4, 135.3, 131.3, 129.8, 129.3, 128.4, 128.2,

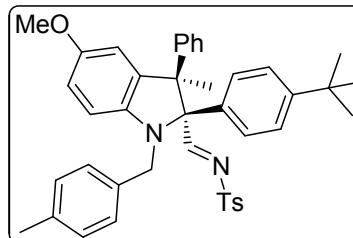
128.0, 127.8, 127.3, 126.4, 126.2, 113.2, 111.3, 109.2, 85.5, 57.8, 55.9, 50.0, 21.7, 21.2, 21.1, 21.0; HRMS: calcd. for  $C_{39}H_{38}N_2O_3S+H$ : 617.2676; found: 617.2694.

**Indoline (3ga):** According to general procedure the product was isolated 73% yield. White solid;  $R_f$  =



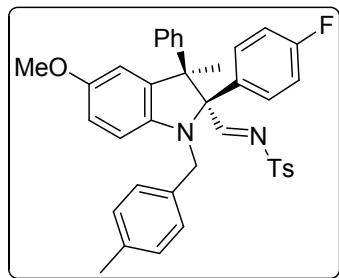
0.68 in 20% ethyl acetate/hexane; mp: 175-177 °C; FTIR (KBr): 3056, 2980, 2928, 1603, 1485, 1449, 1326, 1265, 1160, 1090, 1023, 984, 897, 812, 741, 706, 672, 550  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  8.87 (s, 1H), 7.68 (d, 2H,  $J$  = 8.2 Hz), 7.25 (d, 2H,  $J$  = 8.0 Hz), 7.11-7.09 (m, 4H), 6.98-6.85 (m, 5H), 6.85-6.74 (m, 4H), 6.76 (d, 2H,  $J$  = 7.8 Hz), 6.63-6.56 (m, 4H), 6.16 (d, 1H,  $J$  = 8.4 Hz), 3.99 (ABq, 2H,  $J$  = 16.5, 45.4 Hz), 3.70 (s, 3H), 2.45-2.43 (m, 2H), 2.33 (s, 3H), 1.78 (s, 3H), 1.07 (t, 3H,  $J$  = 7.6 Hz);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  178.7, 154.4, 144.8, 144.7, 143.8, 142.5, 136.2, 136.0, 135.5, 135.3, 131.6, 129.8, 129.3, 128.2, 128.0, 127.8, 127.2, 127.2(2), 126.4, 126.2, 113.2, 111.3, 109.2, 85.5, 58.0, 55.9, 50.0, 28.4, 21.7, 21.1, 21.1(1), 15.7; HRMS: calcd. for  $C_{40}H_{40}N_2O_3S+H$ : 629.2832; found: 629.2834.

**Indoline (3ha):** According to general procedure the product was isolated 66% yield. Colorless liquid;



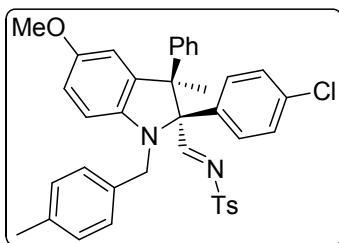
$R_f$  = 0.68 in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 2924, 2858, 1692, 1651, 1598, 1408, 1335, 1172, 1126, 1069, 862, 845, 771, 682, 507, 423  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  8.83 (s, 1H), 7.68 (d, 2H,  $J$  = 8.2 Hz), 7.27 (d, 2H,  $J$  = 8.5 Hz), 7.09-7.12 (m, 4H), 6.97-6.89 (m, 7H), 7.62 (dd, 1H,  $J$  = 2.5, 8.5 Hz), 6.57-6.56 (m, 3H), 6.17 (d, 1H,  $J$  = 8.5 Hz), 4.0 (ABq, 2H,  $J$  = 16.8, 63.2 Hz), 3.71 (s, 3H), 2.44 (s, 3H), 2.34 (s, 3H), 1.77 (s, 3H), 1.66 (s, 9H);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  178.7, 154.4, 150.7, 144.8, 144.7, 142.4, 136.2, 135.9, 135.4, 135.3, 131.4, 129.8, 129.3, 128.2, 128.0, 127.4, 127.2, 126.3, 126.2, 124.6, 113.2, 111.3, 109.2, 85.5, 58.2, 55.9, 50.0, 34.4, 31.3, 21.7, 21.2, 21.0; HRMS: calcd. for  $C_{42}H_{44}N_2O_3S+H$ : 657.3145; found: 657.3129.

**Indoline (3ia):** According to general procedure the product was isolated 92% yield. Colorless liquid;



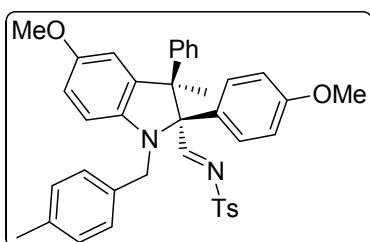
$R_f = 0.68$  in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 2985, 2920, 1602, 1466, 1374, 1245, 1094, 1048, 918, 848, 735, 634, 608  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  8.76 (s, 1H), 7.73 (d, 2H,  $J = 8.3$  Hz), 7.3 (d, 2H,  $J = 7.9$  Hz), 7.18-7.07 (m, 6H), 7.01-6.96 (m, 5H), 6.66-6.59 (m, 4H), 6.21 (d, 1H,  $J = 8.5$  Hz), 3.91 (ABq, 2H,  $J = 16.5, 45.4$  Hz), 3.73 (s, 3H), 2.43 (s, 3H), 2.35 (s, 3H), 1.8 (s, 3H);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  178.2, 163.1(d,  $J = 247.5$  Hz), 154.8, 144.6 (d,  $J = 59.0$  Hz), 136.2 (d,  $J = 44.3$  Hz), 135.3, 134.8, 129.9, 129.9(2), 129.8, 129.7, 129.4, 129.1, 128.3, 127.9, 127.5, 126.6, 126.0, 125.9, 114.4 (d,  $J = 21.4$  Hz), 113.4, 111.3, 109.6, 85.2, 57.5, 55.9, 49.8, 21.7, 21.1, 20.7; HRMS: calcd. for  $\text{C}_{38}\text{H}_{35}\text{FN}_2\text{O}_3\text{S}+\text{Na}$ : 641.2245; found: 641.2269.

**Indoline (3ja):** According to general procedure the product was isolated 85% yield. Colorless liquid;



$R_f = 0.68$  in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 2972, 2829, 1636, 1440, 1375, 1243, 1042, 850, 781, 634, 465, 423  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  8.74 (s, 1H), 7.73 (d, 2H,  $J = 8.3$  Hz), 7.30 (d, 2H,  $J = 8.1$  Hz), 7.12 (d, 2H,  $J = 8.0$  Hz), 7.06 (d, 2H,  $J = 8.0$  Hz), 7.02-6.92 (m, 7H), 6.67-6.59 (m, 4H), 6.21 (d, 1H,  $J = 8.5$  Hz), 3.89 (ABq, 2H,  $J = 16.4, 30.6$  Hz), 3.73 (s, 3H), 2.45 (s, 3H), 2.34 (s, 3H), 1.80 (s, 3H);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  178.0, 154.9, 145.0, 144.3, 142.2, 136.5, 136.0, 135.3, 134.8, 133.6, 132.7, 130.0, 129.9, 129.4, 128.1, 128.1(3), 127.7, 127.5, 126.7, 126.0, 113.4, 111.3, 109.7, 85.3, 57.5, 55.9, 49.9, 21.8, 21.1, 20.7; HRMS: calcd. for  $\text{C}_{38}\text{H}_{35}\text{N}_2\text{ClO}_3\text{S}+\text{H}$ : 635.2130; found: 635.2142.

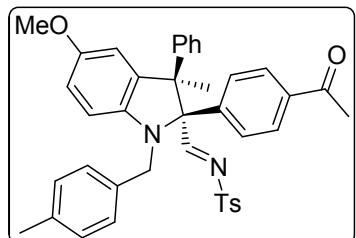
**Indoline (3ka):** According to general procedure the product was isolated 73% yield. Colorless liquid;



$R_f = 0.56$  in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 3055, 2983, 2931, 2836, 1610, 1512, 1491, 1444, 1375, 1327, 1263, 1161, 1090, 1039, 897,

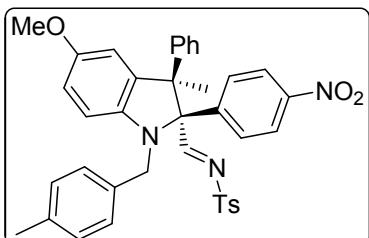
738, 549, 483 cm<sup>-1</sup>; <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>, 24 °C): δ 8.83 (s, 1H), 7.69 (d, 2H, *J* = 8.3 Hz), 7.27 (d, 2H, *J* = 8.0 Hz), 7.18-7.16 (m, 2H), 7.12-7.08 (m, 4H), 6.89-6.88 (m, 1H), 6.64-6.61 (m, 3H), 6.58 (d, 1H, *J* = 2.5 Hz), 6.49 (d, 2H, *J* = 7.8 Hz), 6.17 (d, 2H, *J* = 8.5 Hz), 3.96 (ABq, 2H, *J* = 16.5, 55.1 Hz), 3.72 (s, 3H), 3.65 (m, 3H), 2.44 (s, 3H), 2.34 (s, 3H), 1.78 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C): δ 178.8, 159.0, 154.5, 144.7, 144.7(1), 142.5, 136.2, 136.0, 135.4, 135.3, 129.8, 129.3, 129.1, 128.2, 128.0, 127.3, 126.4, 126.3, 126.1, 113.2, 111.1, 111.3, 109.3, 85.3, 57.8, 55.9, 55.2, 49.9, 21.7, 21.1, 21.0; HRMS: calcd. for C<sub>39</sub>H<sub>38</sub>N<sub>2</sub>O<sub>4</sub>S+H: 631.2625; found: 631.2620.

**Indoline (3la):** According to general procedure the product was isolated 84% yield. Colorless liquid;



R<sub>f</sub> = 0.58 in 20% ethyl acetate/hexane; FTIR (CHCl<sub>3</sub>): 2985, 2920, 1708, 1602, 1506, 1490, 1445, 1374, 1240, 1162, 1092, 1045, 917, 847, 812, 734, 608, 550 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C): δ 8.69 (s, 1H), 7.65 (d, 2H, *J* = 8.3 Hz), 7.45-7.43 (m, 2H), 7.23-7.22 (d, 2H, *J* = 8.0 Hz), 7.10-7.00 (m, 6H), 6.89-6.83 (m, 3H), 6.58 (dd, 1H, *J* = 2.5, 8.5 Hz), 6.54-6.51 (m, 3H), 6.15 (d, 1H, *J* = 8.5 Hz), 3.84 (ABq, 2H, *J* = 16.7, 24.6 Hz), 3.65 (s, 3H), 2.37 (s, 3H), 2.36 (s, 3H), 2.27 (s, 3H), 1.76 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C): δ 197.8, 177.6, 155.0, 145.0, 144.2, 142.0, 139.7, 136.5, 136.0, 135.9, 135.2, 134.7, 130.0, 129.5, 128.3, 128.1, 128.0, 127.5, 127.4, 126.8, 126.0, 113.5, 111.3, 109.8, 85.6, 57.7, 55.9, 50.0, 26.6, 21.8, 21.2, 20.7; HRMS: calcd. for C<sub>40</sub>H<sub>38</sub>N<sub>2</sub>O<sub>4</sub>S+H: 643.2625; found: 643.2642.

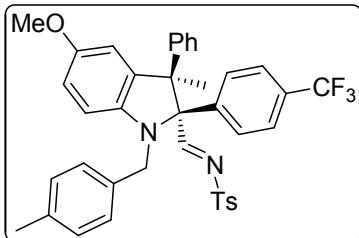
**Indoline (3ma):** According to general procedure the product was isolated 69% yield. Colorless liquid;



R<sub>f</sub> = 0.52 in 20% ethyl acetate/hexane; FTIR (CHCl<sub>3</sub>): 3056, 2984, 2934, 1610, 1520, 1490, 1431, 1346, 1264, 1161, 1091, 1041, 898, 854, 812, 736, 550 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C): δ 8.71 (s, 1H), 7.76 (d, 4H, *J* = 8.3 Hz), 7.33 (d, 2H, *J* = 8.1 Hz), 7.23-7.14 (m, 4H), 7.08 (d, 2H, *J* = 8.0 Hz), 6.99-6.95 (m, 3H), 6.71-6.60 (m, 1H), 6.63-6.59 (m, 3H), 6.29 (d, 1H, *J* = 8.5 Hz), 3.88 (s, 2H), 3.75 (s, 3H), 2.46 (s, 3H), 2.36 (s, 3H), 1.87 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C): δ 176.9, 155.3, 147.1, 145.3,

143.9, 141.8, 141.7, 136.8, 135.8, 135.1, 134.3, 130.1, 129.6, 129.2, 128.2, 127.9, 127.7, 127.1, 125.9, 122.4, 113.8, 111.4, 110.2, 85.6, 57.7, 56.0, 50.0, 21.8, 21.1, 20.6; HRMS: calcd. for  $C_{38}H_{35}N_3O_5S+H$ : 646.2370; found: 646.2371.

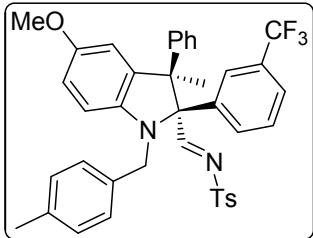
**Indoline (3na):** According to general procedure the product was isolated 77% yield. White solid;  $R_f =$



0.68 in 20% ethyl acetate/hexane; mp: 206-208 °C; FTIR (KBr): 2924, 2834, 1617, 1491, 1411, 1327, 1290, 1242, 1164, 1124, 1089, 1072, 910, 809, 734, 645, 549, 485  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  8.75 (s, 1H), 7.75 (d, 2H,  $J = 8.2$  Hz), 7.32 (d, 2H,  $J = 8.0$  Hz), 7.18-7.07 (m, 8H), 6.99-6.92 (m, 3H), 6.69-6.56 (m, 4H), 6.24 (d, 1H,  $J = 8.4$  Hz), 3.90 (ABq, 2H,  $J = 16.4, 25.6$  Hz), 3.74 (s, 3H), 2.37 (s, 3H), 2.45 (s, 3H), 2.35 (s, 3H), 1.84 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  177.6, 155.1, 145.1, 144.2, 142.0, 138.4, 136.6, 135.8, 135.2, 134.6, 130.0, 129.5, 128.4, 128.1, 128.0, 127.5, 126.8, 126.0, 124.3 (q,  $J = 3.5$  Hz), 113.6, 111.4, 109.9, 85.5, 57.7, 55.9, 49.9, 21.8, 21.1, 20.6; HRMS: calcd. for  $C_{39}H_{35}F_3N_2O_3S+H$ : 669.2393; found: 669.2409.

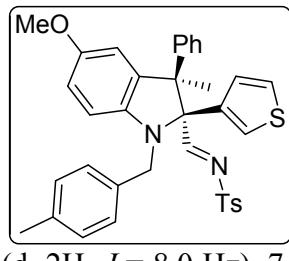
**Indoline (3oa):** According to general procedure the product was isolated 80% yield. Colorless liquid;

Yield: 80%;  $R_f = 0.68$  in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 2973, 2926, 2834, 1614, 1491, 1444,



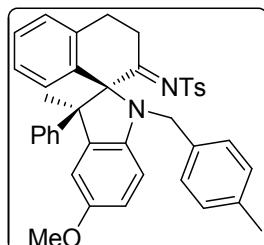
1381, 1288, 1163, 1090, 914, 805, 750, 716, 549, 486  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  8.66 (s, 1H), 7.69 (d, 2H,  $J = 8.0$  Hz), 7.24 (d, 2H,  $J = 8.1$  Hz), 7.20-7.14 (m, 4H), 6.90-6.80 (m, 4H), 6.87-6.85 (m, 3H), 6.61-6.46 (m, 4H), 6.20 (d, 1H,  $J = 8.5$  Hz), 3.85 (s, 2H), 3.65 (s, 3H), 2.37 (s, 3H), 2.26 (s, 3H), 1.74 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  177.3, 155.1, 145.1, 144.2, 142.0, 136.6, 135.8, 135.2, 135.2(6), 134.5, 131.4, 130.0, 129.5, 129.1, 128.3, 128.0, 127.9, 127.9(0), 127.6, 126.8, 126.0, 125.3, 124.2, 124.2(2), 113.6, 111.3, 110.0, 85.3, 58.2, 57.5, 55.9, 49.9, 21.7, 21.1, 20.4; HRMS: calcd. for  $C_{39}H_{35}F_3N_2O_3S+K$ : 707.1952; found: 707.1942.

**Indoline (3pa):** According to general procedure the product was isolated 81% yield. Colorless liquid;



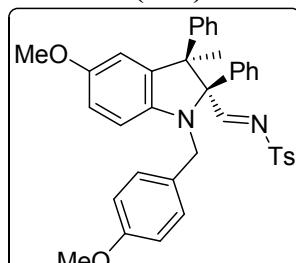
$R_f = 0.58$  in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 3057, 2983, 2938, 1619, 1490, 1375, 1243, 1161, 1091, 1044, 911, 848, 808, 737, 648, 548  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  8.84 (s, 1H), 7.73 (d, 2H,  $J = 8.3$  Hz), 7.30 (d, 2H,  $J = 8.0$  Hz), 7.18-7.12 (m, 4H), 7.07-7.02 (m, 3H), 6.92-6.90 (m, 2H), 6.74-6.62 (m, 4H), 6.59 (d, 1H,  $J = 2.5$  Hz), 6.25 (d, 1H,  $J = 8.4$  Hz), 4.10 (s, 2H), 3.72 (s, 3H), 2.45 (s, 3H), 2.36 (s, 3H), 1.74 (s, 3H);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  178.0, 154.4, 144.8, 144.5, 142.2, 136.3, 136.0, 135.7, 135.4, 135.1, 129.9, 129.3, 128.0, 127.9, 127.5, 127.3, 126.6, 126.2, 124.4, 123.9, 113.2, 111.3, 109.1, 83.9, 58.3, 50.1, 21.7, 21.5, 21.1; HRMS: calcd. for  $\text{C}_{36}\text{H}_{34}\text{N}_2\text{O}_3\text{S}_2+\text{Na}$ : 629.1903; found: 629.1906.

**Indoline (3qa):** According to general procedure the product was isolated 84% yield. White solid;  $R_f =$



0.52 in 20% ethyl acetate/hexane; mp: 95-97 °C; FTIR ( $\text{CHCl}_3$ ): 3058, 2930, 2833, 1605, 1489, 1413, 1346, 1287, 1266, 1159, 1091, 1038, 967, 863, 808, 735, 662, 550  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  7.50 (s, 1H), 7.30 (d, 2H,  $J = 7.7$  Hz), 7.17 (d, 2H,  $J = 7.2$  Hz), 7.04-6.96 (m, 6H), 6.90-6.84 (m, 4H), 6.71-6.69 (m, 1H), 6.62 (d, 2H,  $J = 7.2$  Hz), 6.56-6.55 (m, 1H), 6.37 (dd, 1H,  $J = 1.2, 8.4$  Hz), 6.0-5.99 (m, 1H), 4.22 (d, 1H,  $J = 17.6$  Hz), 3.73 (s, 3H), 3.64 (d, 1H,  $J = 17.4$ ), 2.8 (dd, 1H,  $J = 6.2, 21.3$  Hz), 2.58 (d, 1H,  $J = 21.3$  Hz), 2.42 (s, 3H), 2.31 (s, 3H), 1.98 (s, 3H), 1.53 (s, 2H);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  154.1, 145.1, 143.4, 141.2, 137.1, 136.9, 136.3, 135.9, 135.3, 131.4, 130.0, 129.3, 129.1, 128.3, 127.9, 127.5, 126.9, 126.5, 126.4, 125.8, 125.0, 112.7, 112.4, 108.2, 106.3, 80.6, 61.1, 56.0, 51.1, 29.7, 28.1, 27.0, 21.5, 21.2; HRMS: calcd. for  $\text{C}_{40}\text{H}_{38}\text{N}_2\text{O}_3\text{S}+\text{Na}$ : 649.2495; found: 649.2501.

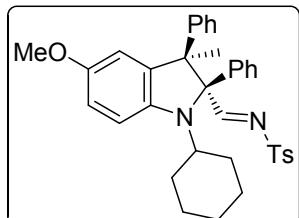
**Indoline (3ab):** According to general procedure the product was isolated 57% yield. Colorless liquid;



$R_f = 0.56$  in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 3057, 2931, 2838,

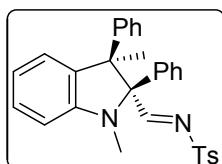
1610, 1491, 1326, 1259, 1162, 1091, 1034, 904, 814, 738, 710, 547 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C): δ 8.79 (s, 1H), 7.62 (d, 2H, *J* = 8.3 Hz), 7.19 (d, 2H, *J* = 8.4 Hz), 7.06 (d, 2H, *J* = 8.8 Hz), 6.97-6.87 (m, 8H), 6.77 (d, 2H, *J* = 8.7 Hz), 6.57-6.53 (m, 3H), 6.50 (d, 1H, *J* = 2.5 Hz), 6.12 (d, 1H, *J* = 8.5 Hz), 3.89 (ABq, 2H, *J* = 16.3, 39.2 Hz), 3.73 (s, 3H), 3.63 (s, 3H), 2.35 (s, 3H), 1.72 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C): δ 178.4, 158.5, 154.5, 144.8, 144.7, 142.3, 136.0, 135.4, 134.5, 130.2, 129.8, 128.2, 128.0, 127.9, 127.7, 127.6, 127.4, 127.3, 126.5, 114.1, 113.3, 111.3, 109.3, 85.5, 58.0, 55.9, 55.3, 49.8, 21.7, 21.1; HRMS: calcd. for C<sub>38</sub>H<sub>36</sub>N<sub>2</sub>O<sub>4</sub>S+Na: 639.2288; found: 639.2292.

**Indoline (3ac):** According to general procedure the product was isolated 34% yield. Colorless liquid;



R<sub>f</sub> = 0.68 in 20% ethyl acetate/hexane; FTIR (CHCl<sub>3</sub>): 3054, 2984, 2928, 2855, 1603, 1487, 1378, 1322, 1265, 1160, 1091, 1038, 909, 739, 650, 550, 436, 416 cm<sup>-1</sup>; <sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>, 24 °C): δ 8.59 (s, 1H), 7.77 (d, 2H, *J* = 8.3 Hz), 7.28 (d, 2H, *J* = 8.0 Hz), 7.24-7.02 (m, 4H), 6.90-6.76 (m, 6H), 6.55 (d, 1H, *J* = 2.6 Hz), 6.34-6.32 (m, 2H), 3.75 (s, 3H), 2.67-2.61 (m, 1H), 2.41 (s, 3H), 2.13-1.90 (m, 2H), 1.78 (s, 3H), 1.74-1.70 (m, 2H), 1.48-1.47 (m, 2H), 1.14-0.98 (m, 4H), 0.66-0.60 (m, 1H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C): δ 176.9, 153.7, 144.7, 143.8, 141.6, 136.5, 135.1, 134.3, 129.8, 128.7, 128.3, 128.1, 127.9, 127.1, 127.1, 126.0, 113.5, 111.8, 110.2, 85.3, 57.0, 56.4, 56.1, 33.6, 31.2, 28.2, 27.9, 27.1, 26.5, 21.7, 19.9; HRMS: calcd. for C<sub>36</sub>H<sub>38</sub>N<sub>2</sub>O<sub>3</sub>S+H: 579.2676; found: 579.2687.

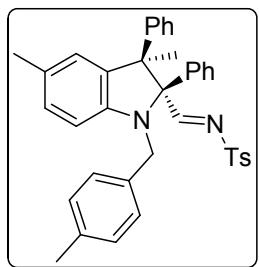
**Indoline (3ad):** According to general procedure the product was isolated 75% yield. White solid; R<sub>f</sub> =



0.68 in 20% ethyl acetate/hexane; mp: 174-176 °C; FTIR (KBr): 3059, 2974, 2929, 1607, 1483, 1323, 1266, 1160, 1091, 1026, 986, 909, 808, 652, 549 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C): δ 8.76 (s, 1H), 7.73 (d, 2H, *J* = 8.3 Hz), 7.29-7.26 (m, 3H), 7.07-6.82 (m, 10H), 6.62 (d, 1H, *J* = 7.8 Hz), 6.47 (d, 2H, *J* = 7.6 Hz), 2.61 (s, 3H), 2.42 (s, 3H), 1.77 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C): δ 177.5, 151.3, 144.8, 143.1, 135.3, 134.3,

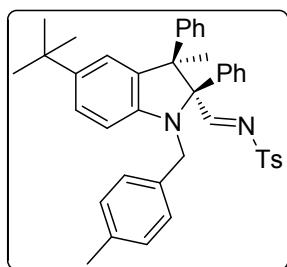
134.0, 129.9, 129.1, 128.3, 128.1, 128.0, 127.5, 127.4, 127.2, 126.2, 124.2, 119.8, 107.1, 84.4, 57.7, 31.4, 21.7, 21.1; HRMS: calcd. for  $C_{30}H_{28}N_2O_2S+Na$ : 503.1764; found: 503.1742.

**Indoline (3af):** According to general procedure the product was isolated 73% yield. Colorless liquid;



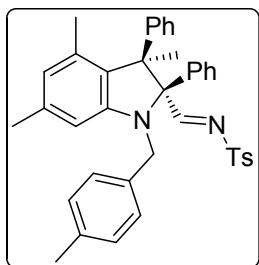
$R_f = 0.62$  in 20% ethyl acetate/hexane; FTIR ( $CHCl_3$ ): 3054, 2975, 2925, 2851, 1613, 1493, 1448, 1330, 1269, 1160, 1090, 1027, 807, 737, 549, 482  $cm^{-1}$ ;  $^1H$  NMR (400 MHz,  $CDCl_3$ , 24 °C):  $\delta$  8.91 (s, 1H), 7.69 (d, 2H,  $J = 7.5$  Hz), 7.28 (d, 2H,  $J = 8.3$  Hz), 7.13-7.11 (m, 4H), 7.05-6.89 (m, 10H), 6.78 (s, 1H), 6.66 (d, 2H,  $J = 7.9$  Hz), 6.21 (d, 1H,  $J = 7.9$  Hz), 4.03 (ABq, 2H,  $J = 16.6, 37.8$  Hz), 2.45 (s, 3H), 2.36 (s, 3H), 2.26 (s, 3H), 1.82 (s, 3H);  $^{13}C\{^1H\}$  NMR (100 MHz,  $CDCl_3$ , 24 °C):  $\delta$  178.3, 148.5, 144.7, 142.6, 136.2, 135.4, 135.2, 134.8, 134.5, 129.8, 129.6, 129.3, 129.0, 128.2, 128.0, 127.9, 127.6, 127.5, 127.3, 126.4, 126.2, 124.9, 108.8, 85.5, 57.9, 49.9, 21.7, 21.3, 21.2, 21.0; HRMS: calcd. for  $C_{38}H_{36}N_2O_2S+Na$ : 607.2390; found: 607.2398.

**Indoline (3ag):** According to general procedure the product was isolated 72% yield. Colorless liquid;



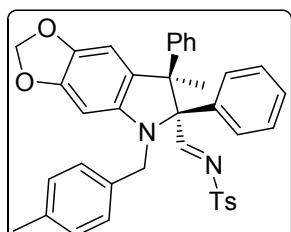
Yield:  $R_f = 0.62$  in 20% ethyl acetate/hexane; FTIR ( $CHCl_3$ ): 3053, 2963, 1613, 1493, 1448, 1332, 1265, 1162, 1088, 903, 741, 545  $cm^{-1}$ ;  $^1H$  NMR (400 MHz,  $CDCl_3$ , 24 °C):  $\delta$  8.79 (s, 1H), 7.62 (d, 2H,  $J = 8.2$  Hz), 7.19-7.14 (m, 3H), 7.03-6.99 (m, 5H), 6.96-6.84 (m, 8H), 6.53 (d, 2H,  $J = 8.1$  Hz), 6.13 (d, 2H,  $J = 8.1$  Hz), 3.91 (ABq, 2H,  $J = 16.6, 34.8$  Hz), 2.34 (s, 3H), 2.26 (s, 3H), 1.75 (s, 3H), 1.16 (s, 9H);  $^{13}C\{^1H\}$  NMR (100 MHz,  $CDCl_3$ , 24 °C):  $\delta$  178.3, 148.2, 144.7, 143.4, 142.2, 136.2, 135.3, 135.2, 134.5, 134.2, 129.8, 129.3, 128.2, 128.0, 127.9, 127.6, 127.5, 127.3, 126.3, 126.2, 125.2, 121.2, 108.3, 85.6, 58.0, 49.9, 34.4, 31.7, 21.7, 21.3, 21.2; HRMS: calcd. for  $C_{41}H_{42}N_2O_2S+Na$ : 649.2859; found: 649.2871.

**Indoline (3ah):** According to general procedure the product was isolated 88% yield. Colorless liquid;



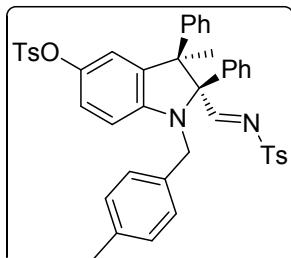
$R_f = 0.58$  in 20% ethyl acetate/hexane; FTIR (CHCl<sub>3</sub>): 3055, 2983, 2927, 2856, 1599, 1481, 1426, 1329, 1266, 1154, 1119, 1023, 975, 908, 739, 522 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C):  $\delta$  8.93 (s, 1H), 7.70 (d, 2H,  $J$  = 8.3 Hz), 7.28 (d, 2H,  $J$  = 8.0 Hz), 7.12-7.07 (m, 4H), 7.03-6.92 (m, 8H), 6.66 (d, 2H,  $J$  = 8.4 Hz), 6.46 (s, 1H), 6.03 (s, 1H), 4.03 (ABq, 2H,  $J$  = 16.8, 33.4 Hz), 2.45 (s, 3H), 2.36 (s, 3H), 2.22 (s, 3H), 1.90 (s, 3H), 1.88 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C):  $\delta$  177.8, 151.1, 144.7, 141.8, 138.5, 135.9, 135.5, 135.3, 134.7, 134.7(1), 129.8, 129.2, 129.1, 128.8, 128.2, 128.0, 127.5, 127.4, 127.3, 126.3, 126.2, 123.8, 107.4, 85.3, 58.3, 50.1, 21.7, 21.6(8), 21.6, 21.1, 19.2; HRMS: calcd. for C<sub>39</sub>H<sub>39</sub>N<sub>2</sub>O<sub>2</sub>S+H: 599.2727; found: 599.2751.

**Indoline (3ai):** According to general procedure the product was isolated 75% yield. Colorless liquid;



$R_f = 0.58$  in 20% ethyl acetate/hexane; FTIR (CHCl<sub>3</sub>): 3056, 2973, 2921, 2883, 1614, 1474, 1375, 1328, 1238, 1188, 1091, 936, 810, 764, 741, 697, 547, 481, 419 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C):  $\delta$  8.92 (s, 1H), 7.72 (d, 2H,  $J$  = 8.1 Hz), 7.29 (d, 2H,  $J$  = 8.4 Hz), 7.13-7.08 (m, 4H), 7.06-6.89 (m, 8H), 6.63 (d, 2H,  $J$  = 7.6 Hz), 6.47 (s, 1H), 5.93 (s, 1H), 5.88 (s, 2H), 3.96 (ABq, 2H,  $J$  = 16.6, 35.0 Hz), 2.45 (s, 3H), 2.34 (s, 3H), 1.77 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C):  $\delta$  178.5, 147.7, 145.7, 144.9, 142.5, 141.4, 136.4, 135.2, 134.8, 134.2, 129.9, 129.4, 129.1, 128.3, 128.1, 128.0, 127.8, 127.7, 127.4, 126.5, 126.1, 105.4, 101.0, 92.9, 85.8, 57.4, 50.0, 21.8, 21.2, 21.1; HRMS: calcd. for C<sub>38</sub>H<sub>34</sub>N<sub>2</sub>O<sub>4</sub>S+H: 615.2312; found: 615.2319.

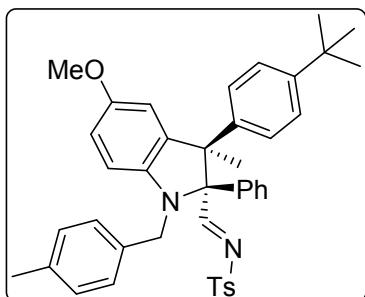
**Indoline (3aj):** According to general procedure the product was isolated 73% yield. Colorless liquid;



$R_f = 0.68$  in 9:91 ethyl acetate/hexane; FTIR (CHCl<sub>3</sub>): 3057, 2978, 2924, 2857, 1604, 1484, 1446, 1371, 1330, 1266, 1161, 1025, 970, 888, 815, 658, 550, 482 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C):  $\delta$  8.96 (s, 1H), 7.65 (d, 2H,  $J$  = 8.3 Hz), 7.59 (d, 2H,  $J$  = 8.3 Hz), 7.27 (d, 2H,  $J$  = 8.0 Hz), 7.23-7.16 (m, 4H),

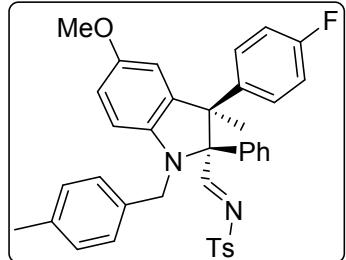
7.13-6.88 (m, 10H), 6.83 (dd, 1H,  $J$  = 2.4, 8.5 Hz), 6.51 (d, 2H,  $J$  = 7.2 Hz), 6.26 (d, 1H,  $J$  = 2.4 Hz), 6.16 (d, 1H,  $J$  = 8.5 Hz), 4.06 (s, 2H), 2.43 (s, 3H), 2.35 (s, 6H), 1.62 (s, 3H);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  176.7, 149.7, 145.2, 145.1, 143.0, 141.4, 136.5, 135.4, 134.9, 134.6, 134.5, 132.3, 129.9, 129.7, 129.4, 129.1, 128.8, 128.0, 127.9, 127.6, 127.5, 126.8, 126.1, 125.4, 123.0, 118.6, 108.6, 85.6, 58.3, 50.2, 21.8, 21.7, 21.5, 21.2; HRMS: calcd. for  $\text{C}_{44}\text{H}_{40}\text{N}_2\text{O}_5\text{S}_2+\text{Na}^+$ : 763.2271; found: 763.2292.

**Indoline (3al):** According to general procedure the product was isolated 81% yield. Colorless liquid;



Yield: 81%;  $R_f$  = 0.68 in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 3056, 2961, 2871, 1612, 1488, 1327, 1267, 1161, 1091, 1035, 810, 733, 552 cm $^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  8.79 (s, 1H), 7.60 (d, 2H,  $J$  = 8.3 Hz), 7.17 (d, 2H,  $J$  = 8.0 Hz), 7.05-7.02 (m, 4H), 6.94-6.80 (m, 7H), 6.55-6.51 (m, 2H), 6.46 (d, 2H,  $J$  = 8.7 Hz), 6.09 (d, 1H,  $J$  = 8.3 Hz), 3.91 (ABq, 2H,  $J$  = 16.4, 27.6 Hz), 3.62 (s, 3H), 2.33 (s, 3H), 2.26 (s, 3H), 1.70 (s, 3H), 1.09 (s, 9H);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  178.7, 154.4, 149.6, 144.7, 144.6, 139.1, 136.2, 136.1, 135.4, 135.2, 134.6, 129.8, 129.3, 128.0, 127.9, 127.7, 127.5, 127.3, 126.2, 124.1, 113.2, 111.3, 109.2, 85.6, 57.9, 55.9, 50.1, 34.2, 31.3, 21.7, 21.2, 21.0; HRMS: calcd. for  $\text{C}_{42}\text{H}_{45}\text{N}_2\text{O}_3\text{S}+\text{H}^+$ : 657.3145; found: 657.3125.

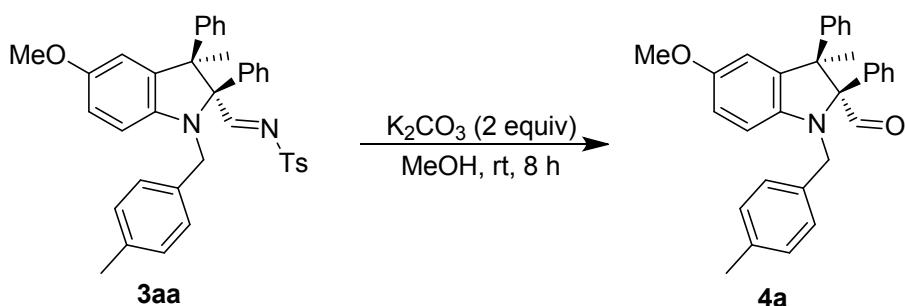
**Indoline (3am):** According to general procedure the product was isolated 60% yield. Colorless liquid;



$R_f$  = 0.68 in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 2984, 2937, 1602, 1507, 1491, 1447, 1374, 1328, 1239, 1162, 1091, 1044, 911, 843, 809, 731, 704, 550 cm $^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  8.84 (s, 1H), 7.71 (d, 2H,  $J$  = 8.3 Hz), 7.29 (d, 2H,  $J$  = 7.9 Hz), 7.15-7.06 (m, 5H), 7.02-6.98 (m, 4H), 6.66-6.54 (m, 6H), 6.21 (d, 1H,  $J$  = 8.5 Hz), 3.99 (ABq, 2H,  $J$  = 16.6, 47.5 Hz), 3.80 (s, 3H), 2.52 (s, 3H), 2.42 (s, 3H), 1.87 (s, 3H);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  178.3, 161.4 (d,  $J$  = 246.4 Hz), 154.7, 144.8, 144.5, 138.3 (d,  $J$  = 3.3 Hz), 136.4, 135.8, 135.3, 135.0, 134.2, 129.9, 129.7,

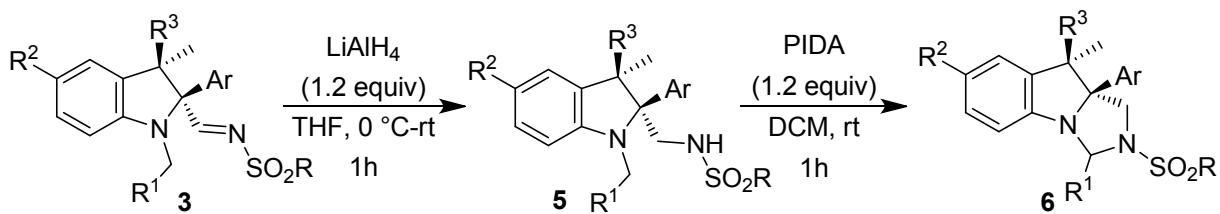
129.4, 128.0, 127.9, 127.8, 127.7, 126.1, 114.0 (d,  $J = 20.5$  Hz), 113.3, 111.2, 109.6, 85.6, 57.3, 55.9, 50.0, 21.7, 21.1, 21.0; HRMS: calcd. for  $C_{38}H_{35}FN_2O_3S+H$ : 619.2425; found: 619.2429.

## 6. Synthesis of compound (**4a**):



Compound **3aa** (100 mg, 0.166 mmol) and  $K_2CO_3$  (0.333 mmol, 2 equiv) were added under nitrogen atmosphere to an oven dried 10 mL reaction tube equipped with stir bar. 4 mL of  $MeOH$  was introduced through syringe and the reaction mixture was stirred at room temperature for 8 h. After the TLC analysis, the reaction mixture was dilute with water (10 mL) and extracted with DCM (2 x 5 mL). The combined organic layer was dried over ( $Na_2SO_4$ ) and filtered. Evaporation of solvent under reduced pressure followed by column chromatography of the crude product afforded the aldehyde **4a** in 69% yield. Colorless liquid;  $R_f = 0.9$  in 20% ethyl acetate/hexane; FTIR ( $CHCl_3$ ): 2989, 2923, 2843, 1702, 1570, 1443, 1327, 1287, 1198, 1105, 986, 862, 636, 412  $cm^{-1}$ ;  $^1H$  NMR (400 MHz,  $CDCl_3$ , 24 °C):  $\delta$  9.97 (s, 1H), 7.27 (d, 2H,  $J = 7.8$  Hz), 7.19 (d, 2H,  $J = 8.0$  Hz), 7.13-7.06 (m, 3H), 7.03-6.97 (m, 5H), 6.70-6.68 (m, 3H), 6.65 (d, 1H,  $J = 2.5$  Hz), 6.31 (d, 1H,  $J = 8.4$  Hz), 4.23 (ABq, 2H,  $J = 16.6$ , 48.7 Hz), 3.74 (s, 3H), 2.38 (s, 3H), 1.98 (s, 3H);  $^{13}C\{^1H\}$  NMR (100 MHz,  $CDCl_3$ , 24 °C):  $\delta$  201.8, 154.4, 145.1, 142.9, 136.6, 136.4, 135.3, 133.9, 129.5, 128.1, 127.9, 127.7, 127.4, 127.3, 126.3, 126.2, 113.1, 111.2, 109.1, 88.2, 56.5, 55.9, 50.2, 21.2, 20.7; HRMS: calcd. for  $C_{31}H_{29}NO_2+H$ : 448.2271; found: 448.2285.

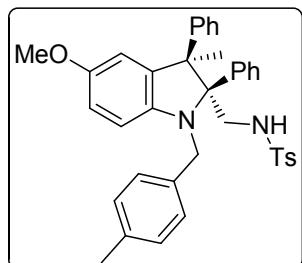
## 7. General procedure for the synthesis of imidazoindolines **6**:



In a 10 mL round bottom flask, compound **3** (1 equiv) was dissolved in dry THF (2 mL) and cooled to 0 °C. After 5 min, LiAlH<sub>4</sub> (1.2 equiv) was added portion-wise to the stirred solution at 0 °C. After completion of addition of LiAlH<sub>4</sub>, the reaction mixture was allowed to warm up to room temperature and stirred for 1 hour. After the consumption of starting material, monitored by TLC analysis, the reaction mixture was quenched by addition of saturated Na<sub>2</sub>SO<sub>4</sub> and extracted with DCM (2 × 5 mL). Organic layers were dried over Na<sub>2</sub>SO<sub>4</sub> followed by evaporation of solvent to give the compound **5**, which was subjected for the next step without further purification.

In a 10 mL reaction tube, compound **5** (obtained above) was dissolved in dry DCM (1 mL). Diacetoxymethane (PIDA, 1.2 equiv) was added at room temperature and the reaction mixture was stirred for 1 hour at same temperature. After the consumption of starting material, monitored by TLC analysis, solvent was removed under the reduced pressure and the crude product was purified by the column chromatography using ethyl acetate and hexane to afford the compound **6** with high yield and purity.

**Compound (5a):** According to the above reduction procedure the product was isolated 90%

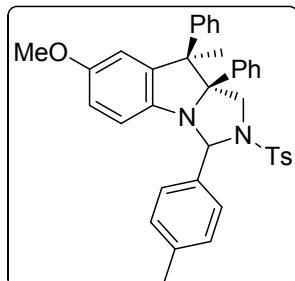


yield. Colorless solid; R<sub>f</sub> = 0.58 in 20% ethyl acetate/hexane; mp: 196-198 °C; FTIR (KBr): 3275, 3092, 2972, 2823, 1586, 1473, 1326, 1289, 1097, 945, 880, 767, 643, 517 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C): δ 7.55 (d, 2H, J = 8.2 Hz), 7.27 (d, 2H, J = 8.0 Hz), 7.16-7.11 (m, 4H), 7.03-6.99 (m, 1H), 6.96-6.87 (m, 5H), 6.73 (d, 1H, J = 7.7 Hz), 6.63 (dd, 1H, J = 2.6, 8.4 Hz), 6.59 (d, 1H, J = 2.5 Hz), 6.55 (d, 2H, J = 7.4 Hz), 6.13 (d, 1H, J = 8.4 Hz), 4.31-4.23 (m, 2H), 3.96-3.91 (m, 2H), 3.72 (s, 3H), 3.53 (dd, 1H, J = 3.2, 12.6 Hz), 2.44 (s, 3H), 2.36 (s, 3H), 1.79 (s, 3H); <sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C): δ 153.8, 145.0, 143.7, 143.5, 138.5, 136.7, 136.3, 136.1, 135.8, 129.8, 129.8(2), 129.5, 128.2,

128.0, 127.3, 127.2, 126.1, 126.0, 112.8, 111.1, 108.5, 81.1, 56.9, 55.9, 49.4, 46.2, 21.7, 21.7(1), 21.2; HRMS: calcd. for  $C_{38}H_{38}N_2O_3S+Na$ : 625.2495; found: 625.2501.

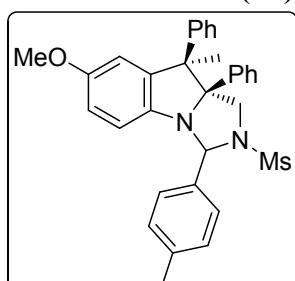
## 8. Properties of isolated imidazoindolines 6

**Imidazoindoline (6a):** According to general procedure the product was isolated 68% yield. White



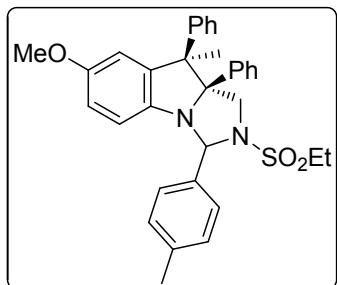
solid;  $R_f = 0.58$  in 20% ethyl acetate/hexane; mp: 195-197 °C; FTIR (KBr): 3107, 2926, 2859, 1600, 1482, 1379, 1344, 1280, 1161, 1097, 1032, 911, 807, 740, 653, 538  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C): 7.41 (d, 2H,  $J = 8.0$  Hz), 7.24 (d, 2H,  $J = 7.8$  Hz), 7.20 (d, 2H,  $J = 8.3$  Hz), 7.00-6.87 (m, 6H), 6.82-6.72 (m, 4H), 6.47 (d, 1H,  $J = 2.6$  Hz), 6.35 (dd, 1H,  $J = 2.6, 8.8$  Hz), 6.21 (d, 2H,  $J = 7.2$  Hz), 5.78 (s, 1H), 5.03 (d, 1H,  $J = 8.9$  Hz), 4.32 (d, 1H,  $J = 10.7$  Hz), 3.97 (d, 1H,  $J = 10.7$  Hz), 3.64 (s, 3H), 2.44 (s, 3H), 2.28 (s, 3H), 1.85 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  155.4, 144.4, 143.9, 142.9, 141.9, 139.1, 138.4, 137.5, 135.6, 132.6, 129.8, 129.2, 128.8, 127.9, 127.4, 127.3, 127.0, 126.5, 125.7, 116.6, 112.9, 109.9, 84.8, 80.7, 55.6, 54.2, 53.4, 21.5, 21.5, 20.0; HRMS: calcd. for  $C_{38}H_{36}N_2O_3S+K$ : 639.2078; found: 639.2059.

**Imidazoindoline (6b):** According to general procedure the product was isolated 74% yield. Colorless



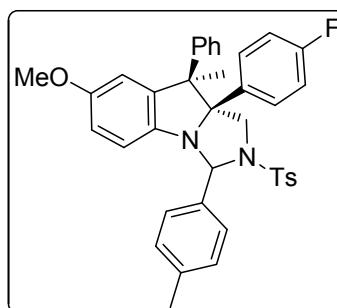
liquid;  $R_f = 0.48$  in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 3052, 2954, 2926, 2858, 1598, 1408, 1172, 1126, 1069, 862, 771, 682, 443  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  7.56-7.51 (m, 1H), 7.42 (d, 2H,  $J = 8.0$  Hz), 7.35-7.31 (m, 1H), 7.23 (d, 2H,  $J = 7.8$  Hz), 7.12-7.04 (m, 3H), 6.99-6.96 (m, 1H), 6.88-6.82 (m, 3H), 6.50 (d, 1H,  $J = 2.6$  Hz), 6.38-6.35 (m, 3H), 5.78 (s, 1H), 5.04 (d, 1H,  $J = 8.9$  Hz), 4.47 (d, 1H,  $J = 11.1$  Hz), 3.97 (d, 1H,  $J = 11.2$  Hz), 3.66 (s, 3H), 2.44 (s, 3H), 2.13 (s, 3H), 1.96 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  155.4, 144.3, 141.6, 139.4, 138.3, 138.2, 132.6, 129.6, 129.1, 128.2, 127.8, 127.5, 127.5(2), 127.2, 125.9, 116.5, 113.0, 109.8, 85.6, 80.1, 55.6, 54.4, 53.8, 37.8, 21.5, 20.1; HRMS: calcd. for  $C_{32}H_{32}N_2O_3S+H$ : 525.2206; found: 525.2214.

**Imidazoindoline (6c):** According to general procedure the product was isolated 68% yield Colorless



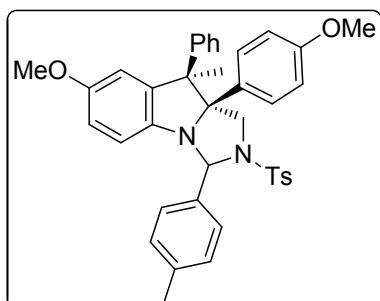
liquid;  $R_f = 0.48$  in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 3054, 3028, 2933, 1598, 1482, 1333, 1278, 1211, 1144, 1034, 993, 897, 807, 733, 700, 514  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  7.41-7.36 (m, 3H), 7.24-7.21 (m, 3H), 7.13-7.07 (m, 2H), 6.94-6.77 (m, 4H), 6.52 (d, 1H,  $J = 2.6$  Hz), 6.37 (dd, 1H,  $J = 2.7, 8.8$  Hz), 6.33 (d, 2H,  $J = 7.1$  Hz), 5.88 (s, 1H), 5.06 (d, 1H,  $J = 8.8$  Hz), 4.59 (d, 1H,  $J = 10.8$  Hz), 3.87 (d, 1H,  $J = 10.8$  Hz), 3.65 (s, 3H), 2.43 (s, 3H), 2.42-2.32 (m, 2H), 1.93 (s, 3H), 1.06 (t, 3H,  $J = 7.3$  Hz);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  155.3, 144.6, 141.8, 139.5, 138.4, 137.5, 132.6, 129.6, 129.1, 128.3, 128.0, 127.7, 127.5, 127.1, 125.9, 116.5, 113.0, 109.9, 85.6, 80.0, 55.6, 53.9, 53.8, 47.0, 21.5, 20.1, 7.6; HRMS: calcd. for  $\text{C}_{33}\text{H}_{34}\text{N}_2\text{O}_3\text{S}+\text{H}$ : 539.2363; found: 539.2360.

**Imidazoindoline (6d):** According to general procedure the product was isolated 57% yield. Colorless



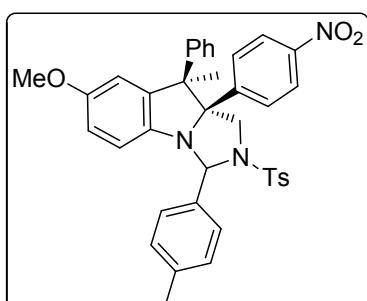
liquid;  $R_f = 0.52$  in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 3072, 2925, 2855, 1597, 1484, 1347, 1277, 1160, 1113, 1092, 902, 809, 782, 735, 699, 597, 539, 459  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  7.41 (d, 2H,  $J = 8.0$  Hz), 7.26 (d, 2H,  $J = 8.0$  Hz), 7.20 (d, 2H,  $J = 7.8$  Hz), 6.94 (d, 2H,  $J = 7.9$  Hz), 6.88-6.76 (m, 6H), 6.58-6.54 (m, 1H), 6.46 (d, 1H,  $J = 2.6$  Hz), 6.35 (dd, 1H,  $J = 2.7, 8.9$  Hz), 6.21 (d, 2H,  $J = 7.1$  Hz), 5.75 (s, 1H), 5.02 (d, 1H,  $J = 8.8$  Hz), 4.27 (d, 1H,  $J = 10.8$  Hz), 3.97 (d, 1H,  $J = 10.8$  Hz), 3.64 (s, 3H), 2.44 (s, 3H), 2.32 (s, 3H), 1.84 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  161.9 (d,  $J = 247.9$  Hz), 155.5, 144.2, 143.4, 141.7, 139.2, 138.2, 135.7, 133.4 (d,  $J = 3.0$  Hz), 132.5, 129.7, 129.3, 128.9, 127.3, 127.2, 126.0, 116.6, 113.3, 114.7 (d,  $J = 21.6$  Hz), 113.0, 109.9, 84.5, 80.7, 55.6, 54.1, 53.4, 21.6, 21.4, 20.0; HRMS: calcd. for  $\text{C}_{38}\text{H}_{35}\text{FN}_2\text{O}_3\text{S}+\text{Na}$ : 641.2245; found: 641.2252.

**Imidazoindoline (6e):** According to general procedure the product was isolated 59% yield. Colorless



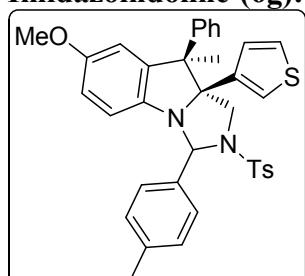
liquid;  $R_f = 0.44$  in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 3023, 2925, 2850, 1603, 1482, 1346, 1252, 1162, 1094, 1032, 810, 736, 698, 666, 588  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  7.40 (d, 2H,  $J = 8.0$  Hz), 7.26-7.24 (m, 2H), 7.19 (d, 2H,  $J = 7.8$  Hz), 6.92 (d, 2H,  $J = 7.9$  Hz), 6.84-6.75 (m, 6H), 6.47 (d, 1H,  $J = 2.6$  Hz), 6.35 (dd, 2H,  $J = 2.7, 8.9$  Hz), 6.21 (d, 2H,  $J = 7.1$  Hz), 5.76 (s, 1H), 5.02 (d, 1H,  $J = 8.8$  Hz), 4.30 (d, 1H,  $J = 10.7$  Hz), 3.94 (d, 1H,  $J = 10.7$  Hz), 3.69 (s, 3H), 3.64 (s, 3H), 2.44 (s, 3H), 2.31 (s, 3H), 1.83 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  158.6, 155.4, 144.5, 143.0, 141.9, 139.1, 138.4, 135.9, 133.5, 132.7, 129.8, 129.4, 129.2, 127.4, 127.4(0), 127.0, 125.8, 125.7, 116.5, 113.3, 112.9, 109.9, 84.5, 80.7, 55.6, 55.2, 54.1, 53.5, 21.6, 21.5, 20.0; HRMS: calcd. for  $\text{C}_{39}\text{H}_{38}\text{N}_2\text{O}_4\text{S}+\text{Na}$ : 653.2444; found: 653.2450.

**Imidazoindoline (6f):** According to general procedure the product was isolated 68% yield. Colorless



liquid;  $R_f = 0.48$  in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 3028, 2925, 2854, 1596, 1505, 1385, 1307, 1280, 1172, 1067, 1017, 931, 862, 770, 611, 416  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  7.74-7.59 (m, 2H), 7.40 (d, 2H,  $J = 8.0$  Hz), 7.33-7.26 (m, 2H), 7.22 (d, 2H,  $J = 7.8$  Hz), 7.13 (d, 2H,  $J = 9.1$  Hz), 6.92 (d, 2H,  $J = 8.0$  Hz), 6.88-6.75 (m, 3H), 6.46 (d, 1H,  $J = 2.6$  Hz), 6.37 (dd, 1H,  $J = 2.7, 8.9$  Hz), 6.25 (d, 2H,  $J = 7.8$  Hz), 5.74 (s, 1H), 5.04 (d, 1H,  $J = 8.9$  Hz), 4.32 (d, 1H,  $J = 11.1$  Hz), 4.03 (d, 1H,  $J = 11.0$  Hz), 3.64 (s, 3H), 2.45 (s, 3H), 2.27 (s, 3H), 1.90 (s, 3H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  155.8, 150.1, 146.5, 146.0, 143.8, 143.4, 141.2, 139.5, 137.8, 135.7, 132.1, 129.6, 129.3, 129.0, 127.5, 127.4, 126.5, 122.9, 116.7, 113.3, 109.8, 84.8, 80.6, 55.6, 54.4, 53.2, 21.6, 21.3, 20.1; HRMS: calcd. for  $\text{C}_{38}\text{H}_{35}\text{N}_3\text{O}_5\text{S}+\text{Na}$ : 668.2190; found: 668.2193.

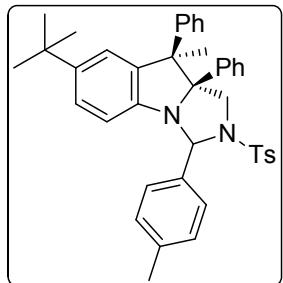
**Imidazoindoline (6g):** According to general procedure the product was isolated 83% yield. Colorless



liquid;  $R_f = 0.52$  in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 3055, 2984,

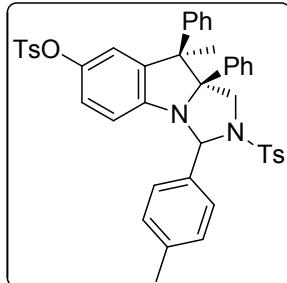
2897, 1597, 1481, 1428, 1344, 1267, 1218, 1161, 1096, 1033, 997, 899, 741, 587  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  7.37 (d, 2H,  $J$  = 8.0 Hz), 7.32 (d, 2H,  $J$  = 8.3 Hz), 7.18 (d, 2H,  $J$  = 7.8 Hz), 7.0 (d, 2H,  $J$  = 8.0 Hz), 6.90-6.82 (m, 3H), 6.75 (dd, 1H,  $J$  = 2.9, 4.9 Hz), 6.66 (dd, 1H,  $J$  = 1.2, 2.8 Hz), 6.49 (d, 1H,  $J$  = 2.7 Hz), 6.44 (dd, 1H,  $J$  = 1.0, 5.0 Hz), 6.35 (dd, 1H,  $J$  = 2.7, 8.9 Hz), 6.29 (d, 2H,  $J$  = 7.2 Hz), 5.83 (s, 1H), 5.02 (d, 1H,  $J$  = 8.9 Hz), 4.21 (d, 1H,  $J$  = 10.5 Hz), 3.94 (d, 1H,  $J$  = 10.5 Hz), 3.65 (s, 3H), 2.44 (s, 3H), 2.34 (s, 3H), 1.84 (s, 3H).  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  155.5, 144.6, 143.1, 141.8, 139.9, 139.1, 138.3, 135.9, 132.4, 129.8, 129.3, 128.8, 127.3, 127.2, 127.1, 126.2, 125.9, 124.8, 122.2, 116.5, 112.9, 109.9, 83.0, 81.2, 55.6, 54.0, 53.8, 21.6, 21.5, 19.9; HRMS: calcd. for  $\text{C}_{36}\text{H}_{35}\text{N}_2\text{O}_3\text{S}_2+\text{Na}$ : 607.2084; found: 607.2098.

**Imidazoindoline (6h):** According to general procedure the product was isolated 89% yield Colorless



liquid;  $R_f$  = 0.56 in 20% ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 2957, 2928, 2866, 1601, 1486, 1348, 1272, 1160, 1093, 998, 895, 808, 739, 699, 673, 581, 519  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  7.45 (d, 2H,  $J$  = 7.8 Hz), 7.26 (d, 2H,  $J$  = 8.1 Hz), 7.21 (d, 2H,  $J$  = 7.8 Hz), 6.94-6.88 (m, 6H), 6.82-6.79 (m, 3H), 6.88-6.82 (m, 3H), 6.18 (d, 2H,  $J$  = 7.3 Hz), 5.80 (s, 1H), 5.05 (d, 1H,  $J$  = 8.6 Hz), 4.32 (d, 1H,  $J$  = 10.8 Hz), 3.97 (d, 1H,  $J$  = 10.7 Hz), 2.46 (s, 3H), 2.29 (s, 3H), 1.87 (s, 3H), 1.17 (s, 9H);  $^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  145.8, 145.1, 144.9, 142.9, 139.0, 137.6, 136.6, 135.6, 132.6, 129.8, 129.2, 128.8, 127.9, 127.5, 127.3, 127.3(3), 126.9, 126.4, 125.5, 124.3, 121.1, 115.1, 84.7, 80.5, 54.2, 53.3, 34.3, 31.6, 21.6, 21.5, 20.2; HRMS: calcd. for  $\text{C}_{41}\text{H}_{42}\text{N}_2\text{O}_2\text{S}+\text{H}$ : 627.3040; found: 627.3051.

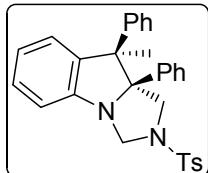
**Imidazoindoline (6i):** According to general procedure the product was isolated 70% yield. Colorless



liquid;  $R_f$  = 0.46 in 16:84 ethyl acetate/hexane; FTIR ( $\text{CHCl}_3$ ): 3024, 2925, 2862, 1597, 1474, 1367, 1270, 1163, 1092, 1002, 918, 817, 762, 703, 661, 552  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  7.51 (d, 2H,  $J$  = 8.2 Hz), 7.35 (d,

2H,  $J = 8.0$  Hz), 7.22 (d, 2H,  $J = 8.2$  Hz), 7.18 (d, 2H,  $J = 7.8$  Hz), 7.07 (d, 2H,  $J = 8.0$  Hz), 6.92-6.82 (m, 8H), 6.77-6.73 (m, 2H), 6.51 (dd, 1H,  $J = 2.5, 8.9$  Hz), 6.27 (d, 1H,  $J = 2.5$  Hz), 6.12 (d, 2H,  $J = 7.5$  Hz), 5.77 (s, 1H), 4.99 (d, 1H,  $J = 8.9$  Hz), 4.32 (d, 1H,  $J = 10.7$  Hz), 3.88 (d, 1H,  $J = 10.7$  Hz), 2.43 (s, 3H), 2.30 (s, 3H), 2.28 (s, 3H), 1.72 (s, 3H);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  146.9, 145.1, 144.5, 143.6, 143.1, 139.5, 138.4, 136.9, 135.4, 132.0, 132.0(1), 129.6, 129.5, 129.3, 129.0, 128.8, 128.6, 128.0, 127.2, 127.1, 126.7, 126.0, 125.9, 121.7, 118.3, 116.0, 84.8, 80.0, 53.8, 53.2, 21.6, 21.5, 21.5, 20.0; HRMS: calcd. for  $\text{C}_{44}\text{H}_{40}\text{N}_2\text{O}_5\text{S}_2+\text{H}$ : 741.2451; found: 741.2458.

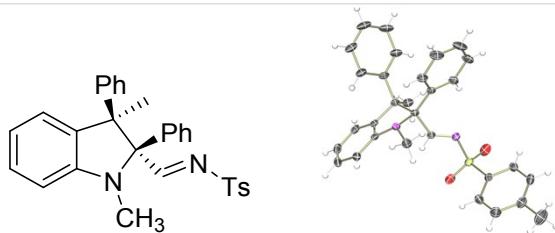
**Imidazoindoline (6k):** According to general procedure the product was isolated 69% yield. White



solid;  $R_f = 0.56$  in 20% ethyl acetate/hexane; mp: 188-190 °C; FTIR (KBr): 3043, 2979, 2925, 1595, 1408, 1335, 1171, 1127, 1069, 862, 771, 682, 412  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  7.40 (d, 2H,  $J = 8.3$  Hz), 7.17-7.13 (m, 1H), 7.0-6.94 (m, 5H), 6.92-6.83 (m, 5H), 6.81-6.77 (m, 2H), 6.88-6.82 (m, 3H), 6.70 (d, 1H,  $J = 6.9$  Hz), 6.34-6.32 (m, 2H), 4.94 (d, 1H,  $J = 8.7$  Hz), 4.32 (d, 1H,  $J = 8.7$  Hz), 3.93 (d, 1H,  $J = 9.6$  Hz), 3.48 (d, 1H,  $J = 9.6$  Hz), 2.31 (s, 3H), 1.83 (s, 3H);  $^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C):  $\delta$  150.5, 149.6, 143.6, 143.3, 137.2, 136.4, 134.1, 129.5, 128.6, 127.9, 127.5, 127.1, 127.0, 126.3, 126.0, 124.5, 122.5, 111.6, 86.4, 66.2, 54.7, 52.1, 21.5, 20.2; HRMS: calcd. for  $\text{C}_{30}\text{H}_{28}\text{N}_2\text{O}_2\text{S}+\text{Na}$ : 503.1764; found: 503.1767.

## 9. Crystallographic data and structure refinements summary for compound 3ad

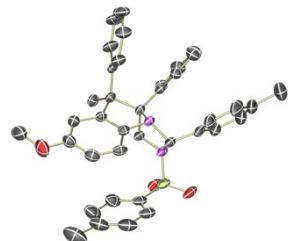
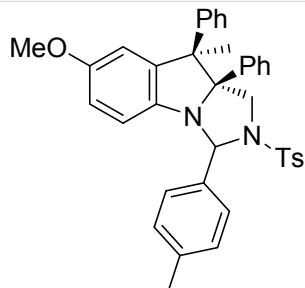
Recrystallization of indoline **3ad** from DCM : hexane (1 : 2) by slow evaporation in a vial with diameter of 0.75 cm and length 5 cm at 30 °C afforded suitable size and quality crystals for X-ray diffraction.

Molecular Structure (Ortep Structure)	
CCDC number	1438964
Formula	C <sub>30</sub> H <sub>28</sub> N <sub>2</sub> O <sub>2</sub> S
Formula weight	480.6
Color	Colorless
Temperature/K	296(2)
Radiation	Mo K $\alpha$
Wavelength/ $\text{\AA}$	0.71073
Crystal system	Monoclinic
Space group	P 21/n
<i>a</i> ( $\text{\AA}$ )	8.5621(3)
<i>b</i> ( $\text{\AA}$ )	12.1149(4)
<i>c</i> ( $\text{\AA}$ )	25.0836(10)
$\alpha$ (°)	90
$\beta$ (°)	99.7094(14)
$\gamma$ (°)	90
Volume ( $\text{\AA}^3$ )	2564.63(16)
<i>Z</i>	4
Density (g/ml)	1.245
$\mu$ (1/mm)	0.156
<i>F</i> (000)	1016
$\theta$ (min, max)	1.647, 24.997
No. of unique reflns	4520
No. of parameters	319
<i>R</i> _obs, <i>wR</i> <sub>2</sub> _obs	0.0398, 0.0961
$\Delta\rho_{\text{min}}, \Delta\rho_{\text{max}}$ (e $\text{\AA}^{-3}$ )	-0.318, 0.199
GooF	1.016

## 10. Crystallographic data and structure refinements summary for compound 6a

Recrystallization of imidazoindoline **6a** from EtOAc : hexane (1 : 2) by slow evaporation in a vial with diameter of 0.75 cm and length 5 cm at 30 °C afforded suitable size and quality crystals for X-ray diffraction.

Molecular Structure (Ortep Structure)

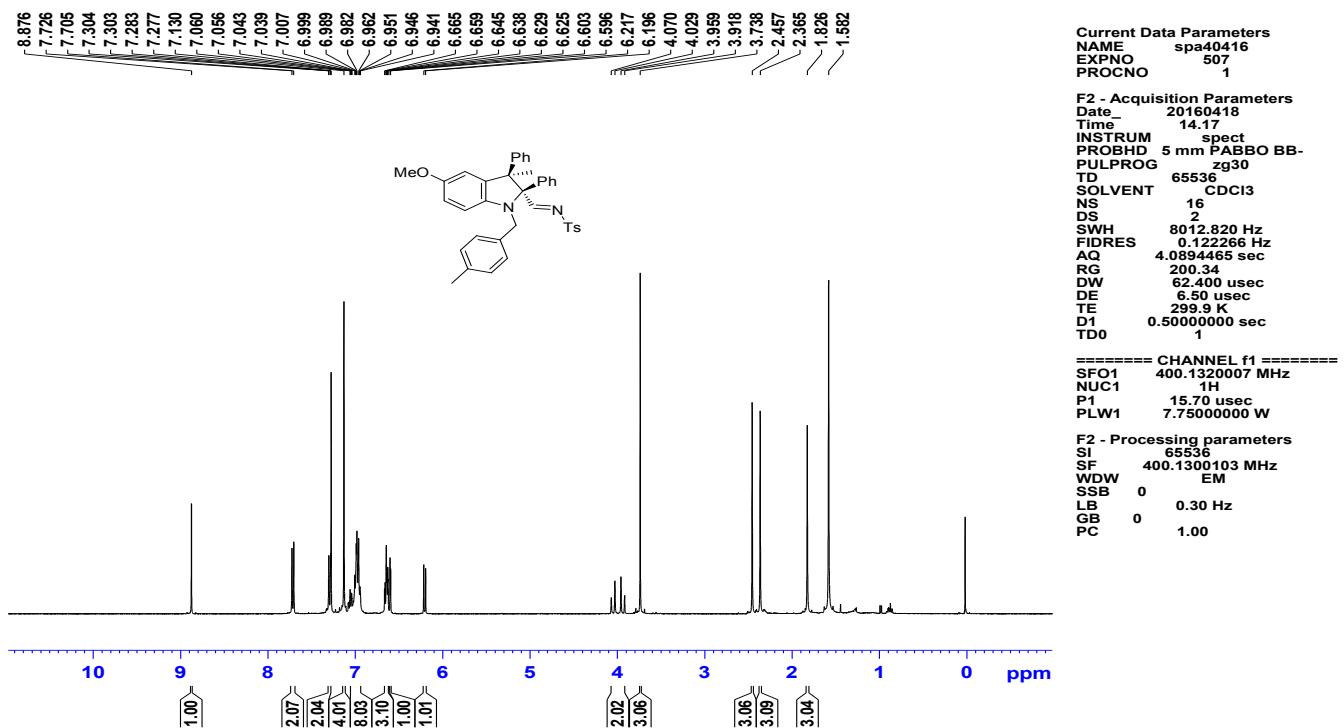


CCDC number	1438963
Formula	C <sub>38</sub> H <sub>36</sub> N <sub>2</sub> O <sub>3</sub> S
Formula weight	600.75
Color	White
Temperature/K	296(2)
Radiation	Mo K $\alpha$
Wavelength/ $\text{\AA}$	0.71073
Crystal system	Monoclinic
Space group	P 21/c
a ( $\text{\AA}$ )	6.8957(3)
b ( $\text{\AA}$ )	18.2571(8)
c ( $\text{\AA}$ )	26.4728(13)
$\alpha$ (°)	90
$\beta$ (°)	91.3638(19)
$\gamma$ (°)	90
Volume ( $\text{\AA}^3$ )	3331.9(3)
Z	4
Density (g/ml)	1.198
$\mu$ (1/mm)	0.135
F (000)	1272
$\theta$ (min, max)	1.539, 24.261
No. of unique reflns	5343
No. of parameters	402
$R_{\text{obs}}$ , $wR_{2\text{-obs}}$	0.0642, 0.1774
$\Delta\rho_{\text{min}}$ , $\Delta\rho_{\text{max}}$ (e $\text{\AA}^{-3}$ )	-0.244, 0.958
GooF	1.063

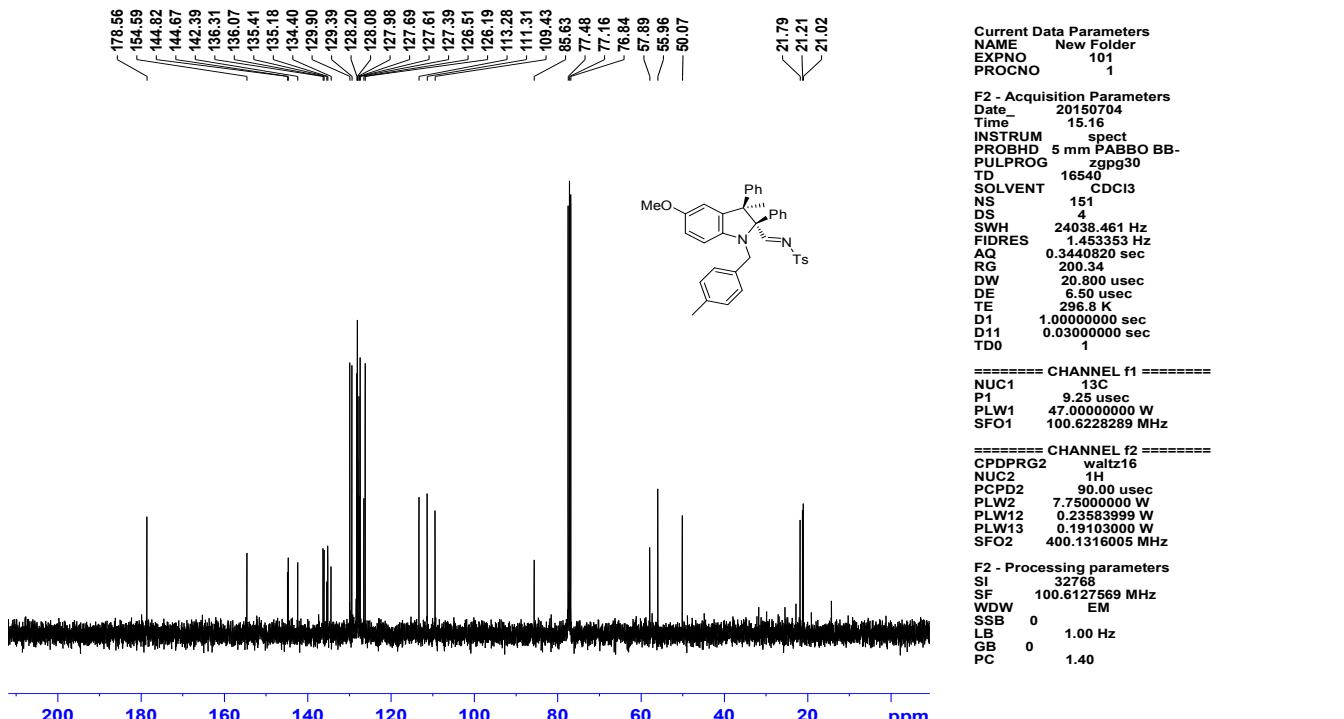
## 11. NMR spectra of isolated compounds:

Indoline (3aa):

<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C)

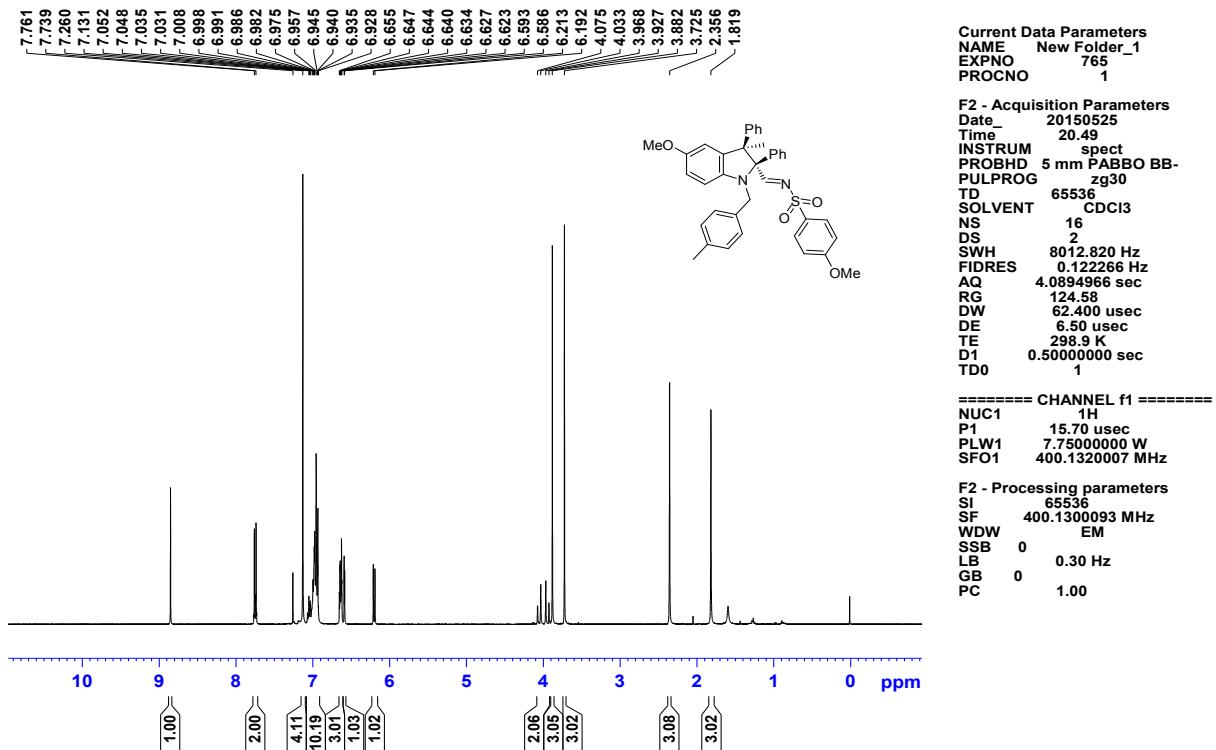


<sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C)

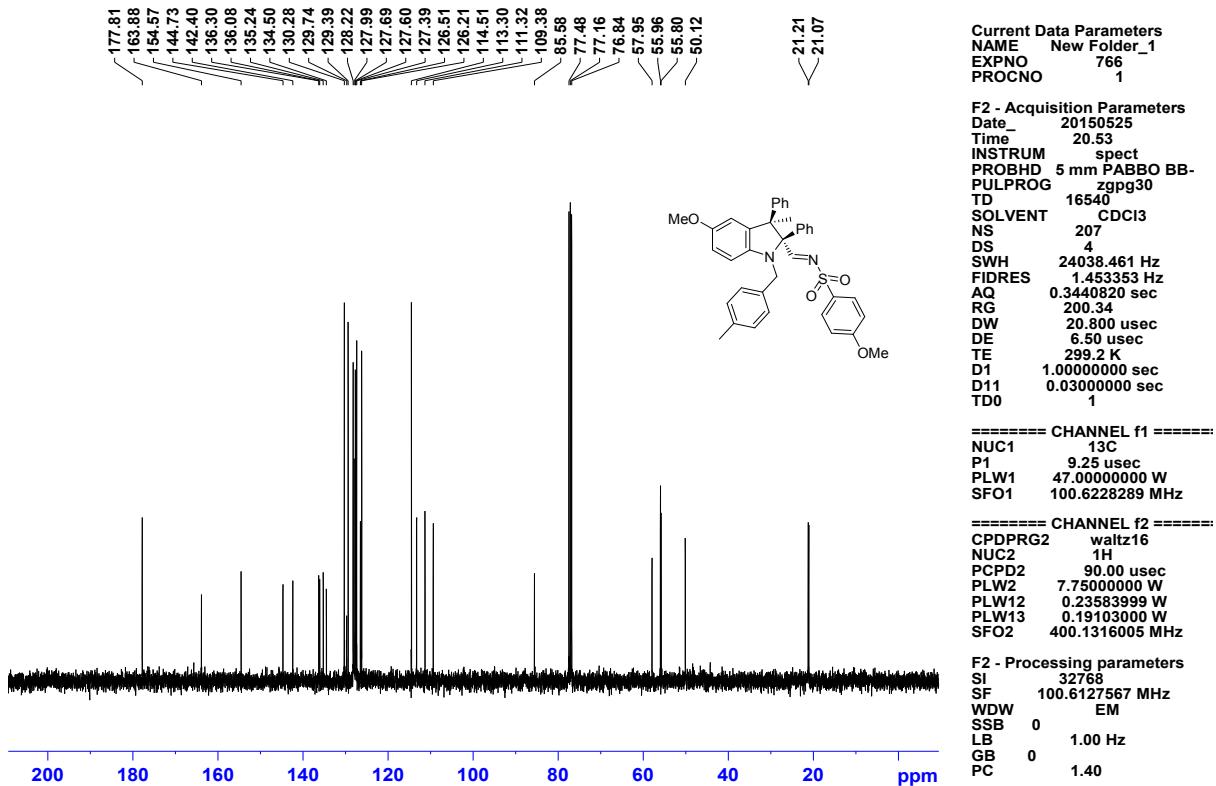


**Indoline (3ba) :**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

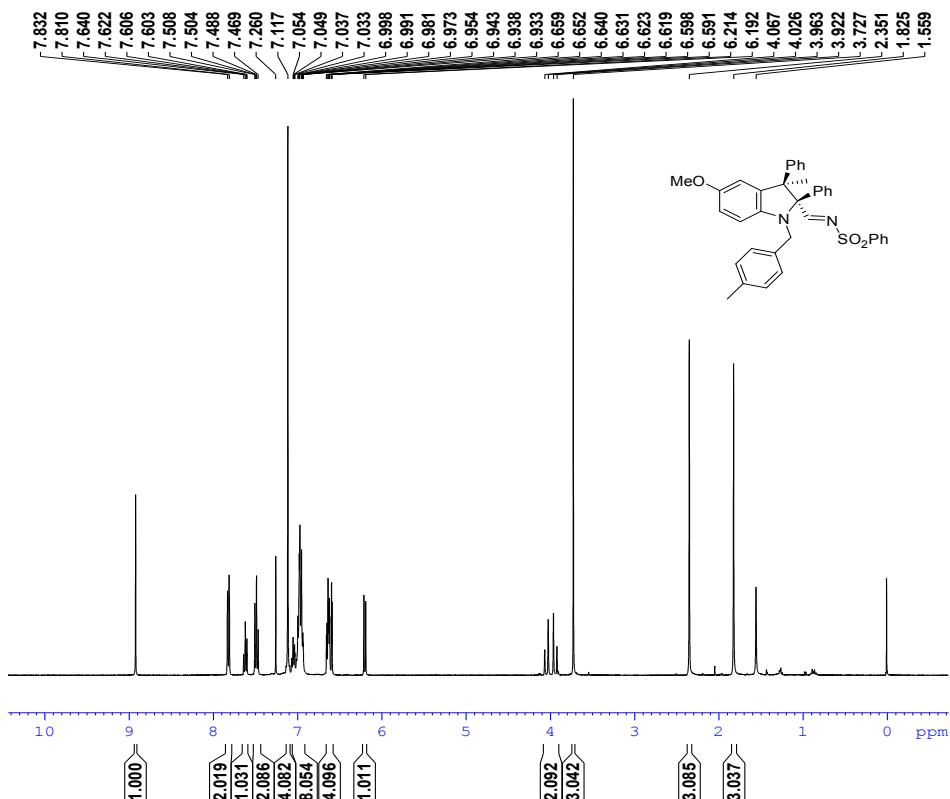


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



**Indoline (3ca):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**



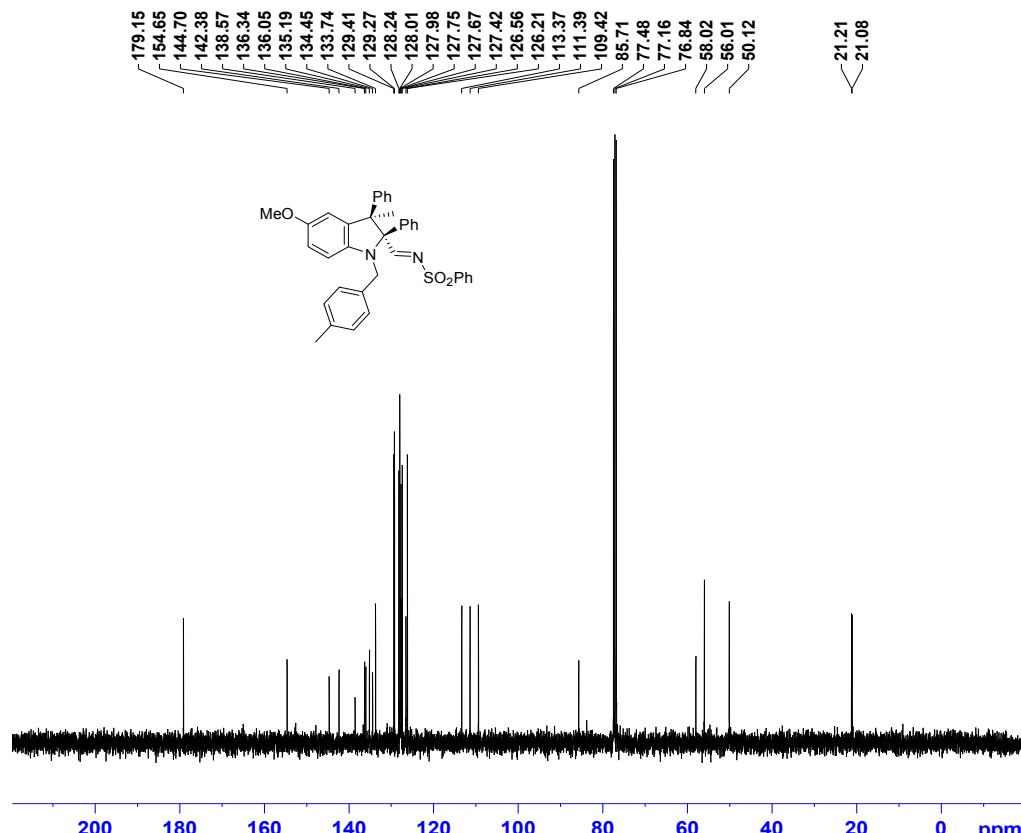
Current Data Parameters  
NAME New Folder\_1  
EXPNO 701  
PROCNO 1

F2 - Acquisition Parameters  
Date 20150522  
Time 1.06  
INSTRUM spect  
PROBHD 5 mm PABBO BB-PULPROG zg30  
TD 65536  
SOLVENT CDCl3  
NS 16  
DS 2  
SWH 8012.820 Hz  
FIDRES 0.122266 Hz  
AQ 4.0894465 sec  
RG 169.77  
DW 62.400 usec  
DE 6.50 usec  
TE 301.2 K  
D1 0.5000000 sec  
TD0 1

===== CHANNEL f1 ======  
NUC1 1H  
P1 15.70 usec  
PLW1 7.7500000 W  
SFO1 400.1320007 MHz

F2 - Processing parameters  
SI 65536  
SF 400.1300094 MHz  
WDW EM  
SSB 0  
LB 0.30 Hz  
GB 0 1.00  
PC 1.00

**$^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



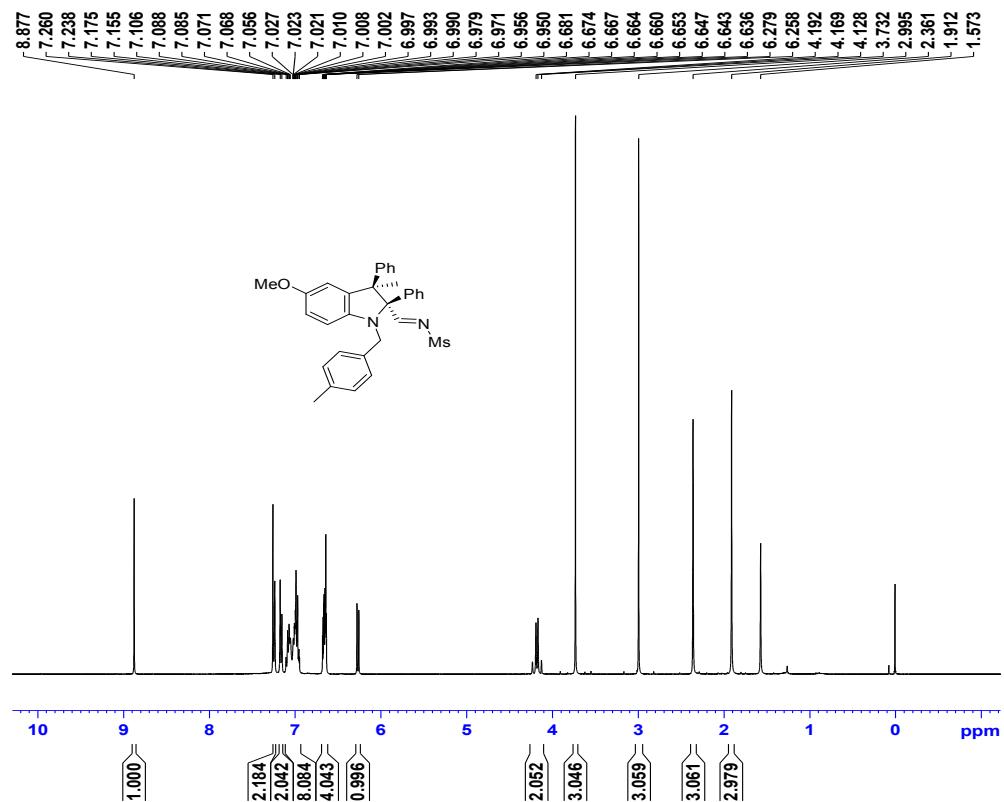
Current Data Parameters  
NAME New Folder\_1  
EXPNO 702  
PROCNO 1

F2 - Acquisition Parameters  
Date 20150522  
Time 1.14  
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PROBHD 5 mm PABBO BB-PULPROG zgpg30  
TD 16540  
SOLVENT CDCl3  
NS 256  
DS 4  
SWH 24038.461 Hz  
FIDRES 1.453353 Hz  
AQ 0.3440320 sec  
RG 200.34  
DW 20.800 usec  
DE 6.50 usec  
TE 302.0 K  
D1 1.0000000 sec  
d11 0.03000000 sec  
DELTA 0.8999998 sec  
TD0 1  
SFO1 100.6228289 MHz  
NUC1 13C  
P1 9.25 usec  
PLW1 47.0000000 W  
SFO2 400.1316005 MHz  
NUC2 1H  
CPDPG[2] waltz16  
PCPD2 90.00 usec  
PLW2 7.7500000 W  
PLW12 0.23583999 W  
PLW13 0.19103000 W

F2 - Processing parameters  
SI 32768  
SF 100.6127541 MHz  
WDW EM  
SSB 0  
LB 1.00 Hz  
GB 0 1.40  
PC

**Indoline (3da):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**



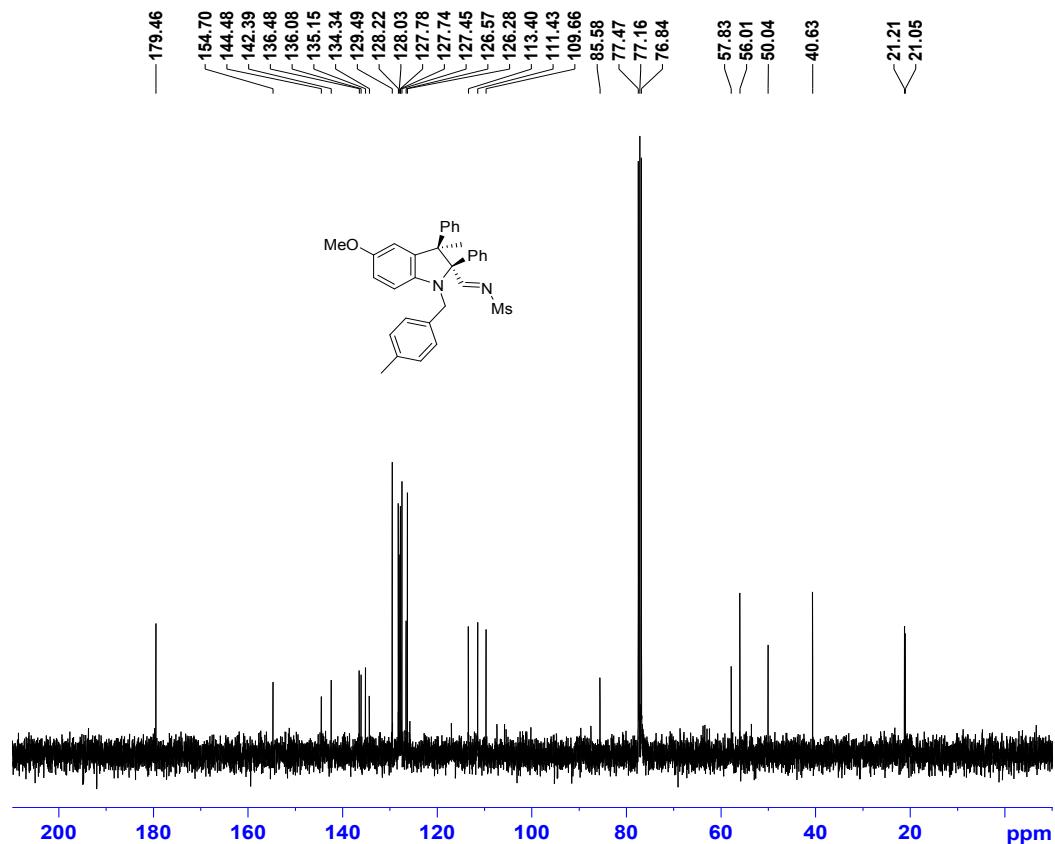
Current Data Parameters  
 NAME spa40415  
 EXPNO 780  
 PROCNO 1

F2 - Acquisition Parameters  
 Date\_ 20150425  
 Time 18.36  
 INSTRUM spect  
 PROBHD 5 mm PABBI 1H/  
 PULPROG zg30  
 TD 65536  
 SOLVENT CDCl3  
 NS 16  
 DS 2  
 SWH 8012.820 Hz  
 FIDRES 0.122266 Hz  
 AQ 4.0894465 sec  
 RG 108.26  
 DW 62.400 usec  
 DE 6.50 usec  
 TE 298.5 K  
 D1 0.5000000 sec  
 TD0 1

===== CHANNEL f1 ======  
 NUC1  $^1\text{H}$   
 P1 9.00 usec  
 PLW1 8.5000000 W  
 SFO1 400.1320007 MHz

F2 - Processing parameters  
 SI 65536  
 SF 400.1300102 MHz  
 WDW EM  
 SSB 0  
 LB 0.30 Hz  
 GB 0  
 PC 1.00

**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



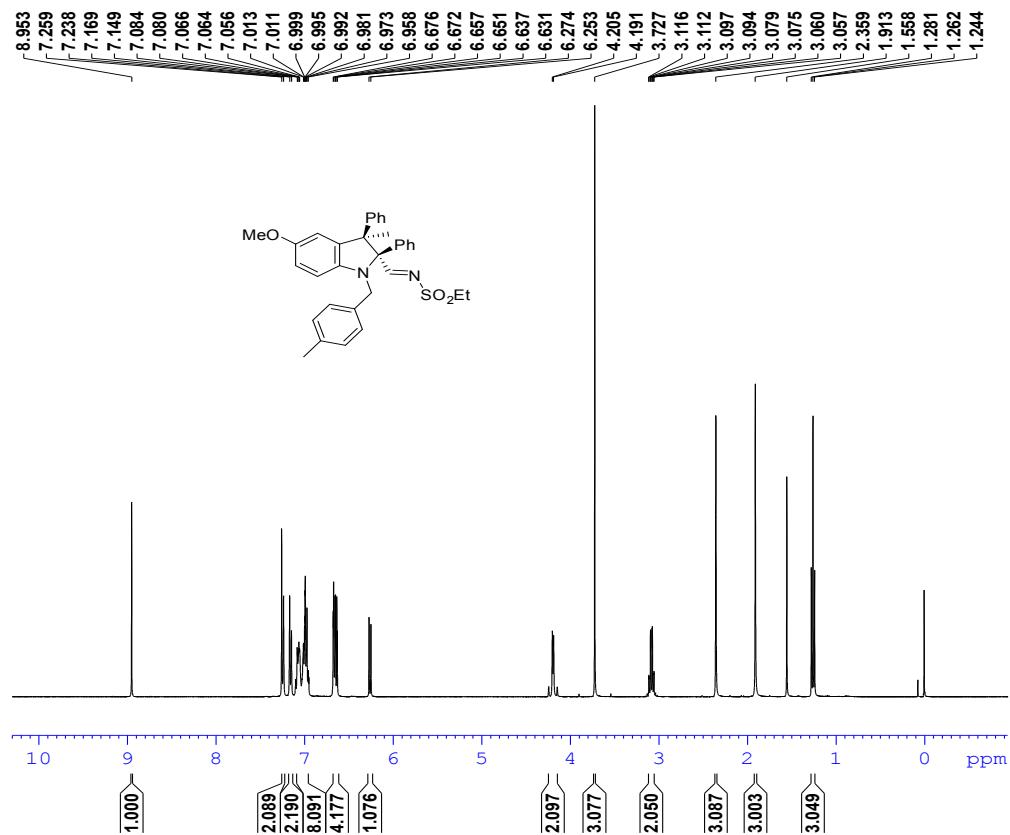
Current Data Parameters  
 NAME spa40415  
 EXPNO 781  
 PROCNO 1

F2 - Acquisition Parameters  
 Date\_ 20150425  
 Time 18.43  
 INSTRUM spect  
 PROBHD 5 mm PABBI 1H/  
 PULPROG zgpg30  
 TD 16540  
 SOLVENT CDCl3  
 NS 256  
 DS 4  
 SWH 24038.461 Hz  
 FIDRES 1.453353 Hz  
 AQ 0.3440320 sec  
 RG 200.34  
 DW 20.800 usec  
 DE 6.50 usec  
 TE 298.7 K  
 D1 1.00000000 sec  
 d11 0.03000000 sec  
 DELTA 0.89999998 sec  
 TD0 1  
 SFO1 100.6228289 MHz  
 NUC1  $^{13}\text{C}$   
 P1 15.00 usec  
 PLW1 76.00000000 W  
 SFO2 400.1316005 MHz  
 NUC2  $^1\text{H}$   
 CPDPRG[2] waltz16  
 PCPD2 90.00 usec  
 PLW2 8.50000000 W  
 PLW12 0.08500000 W  
 PLW13 0.06885000 W

F2 - Processing parameters  
 SI 32768  
 SF 100.6127559 MHz  
 WDW EM  
 SSB 0  
 LB 1.00 Hz  
 GB 0  
 PC 1.40

**Indoline (3ea):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**



Current Data Parameters  
NAME New Folder\_1  
EXPNO 707  
PROCNO 1

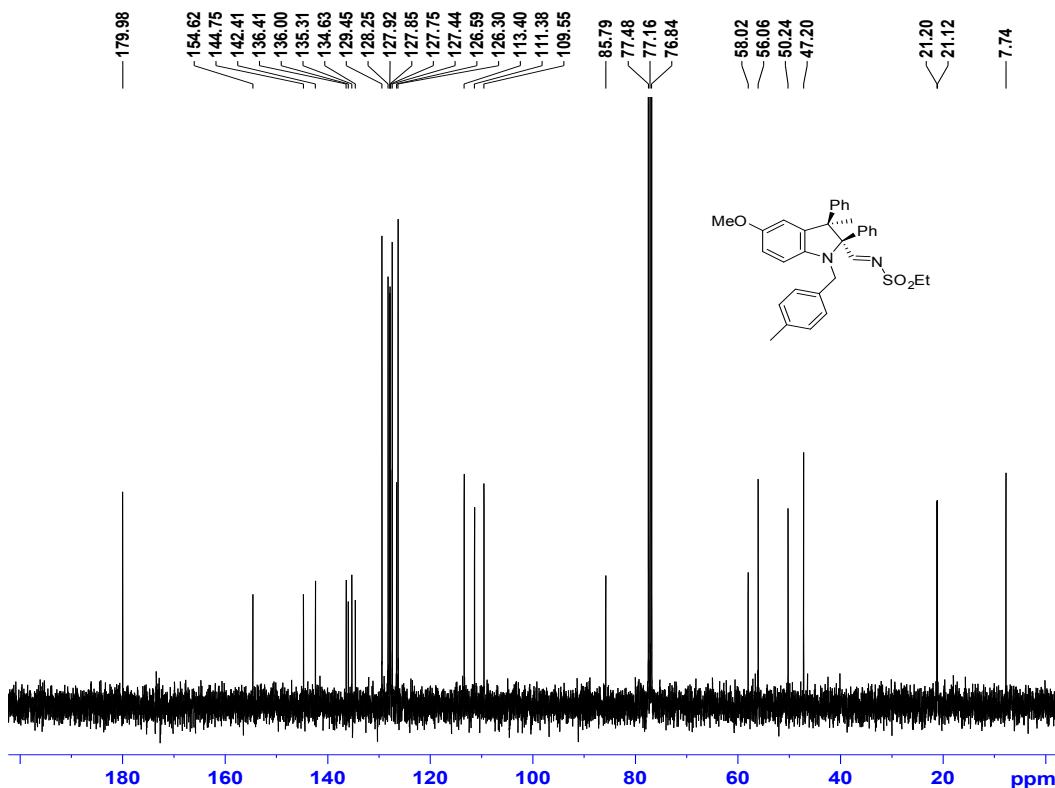
F2 - Acquisition Parameters  
Date 20150522  
Time 1.42  
INSTRUM spect  
PROBHD 5 mm PABBO BB-  
PULPROG zg30  
TD 65536  
SOLVENT CDCl3  
NS 16  
DS 2  
SWH 8012.820 Hz  
FIDRES 0.122266 Hz  
AQ 4.0894465 sec  
RG 169.77  
DW 62.400 usec  
DE 6.50 usec  
TE 301.2 K  
D1 0.5000000 sec  
TD0 1

===== CHANNEL f1 ======

NUC1 1H  
P1 15.70 usec  
PLW1 7.7500000 W  
SFO1 400.1320007 MHz

F2 - Processing parameters  
SI 65536  
SF 400.1300097 MHz  
WDW EM  
SSB 0  
LB 0.30 Hz  
GB 0  
PC 1.00

**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



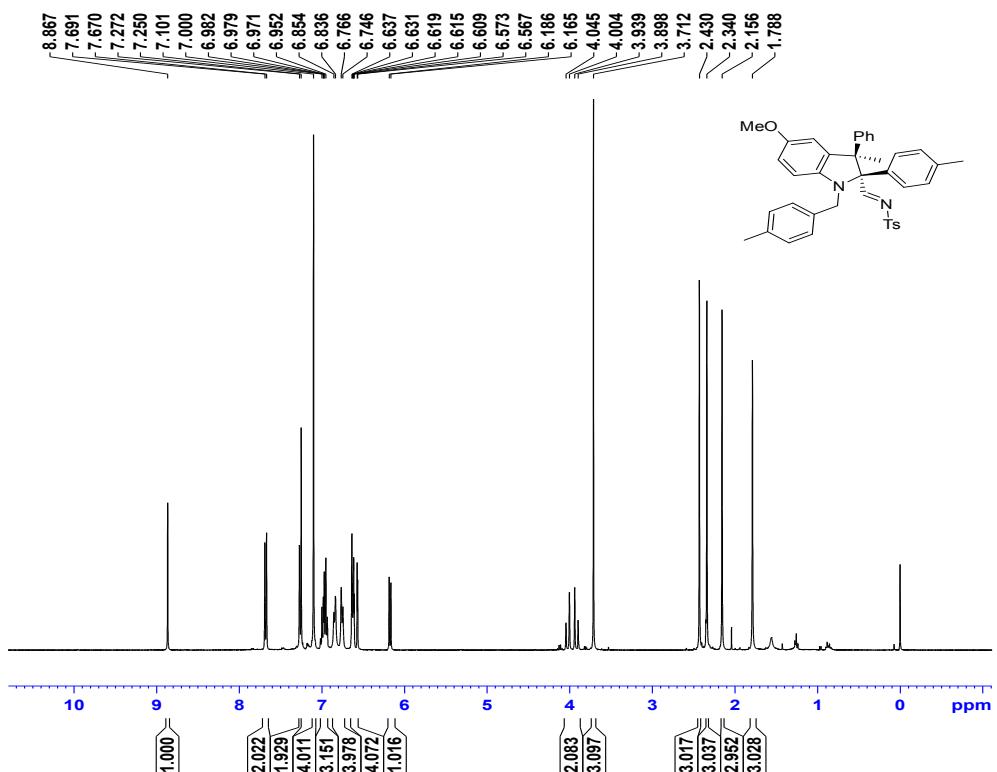
Current Data Parameters  
NAME New Folder\_1  
EXPNO 708  
PROCNO 1

F2 - Acquisition Parameters  
Date 20150522  
Time 1.49  
INSTRUM spect  
PROBHD 5 mm PABBO BB-  
PULPROG zgpg30  
TD 16540  
SOLVENT CDCl3  
NS 256  
DS 4  
SWH 24038.461 Hz  
FIDRES 1.4535353 Hz  
AQ 0.3440320 sec  
RG 200.34  
DW 20.800 usec  
DE 6.50 usec  
TE 302.0 K  
D1 1.000000000 sec  
d11 0.030000000 sec  
DELTA 0.89999998 sec  
TD0 1  
SFO1 100.6228289 MHz  
NUC1 13C  
P1 9.25 usec  
PLW1 47.000000000 W  
SFO2 400.1316005 MHz  
NUC2 1H  
CPDPG[2 waltz16  
PCPD2 90.00 usec  
PLW2 7.750000000 W  
PLW12 0.23583999 W  
PLW13 0.19103000 W

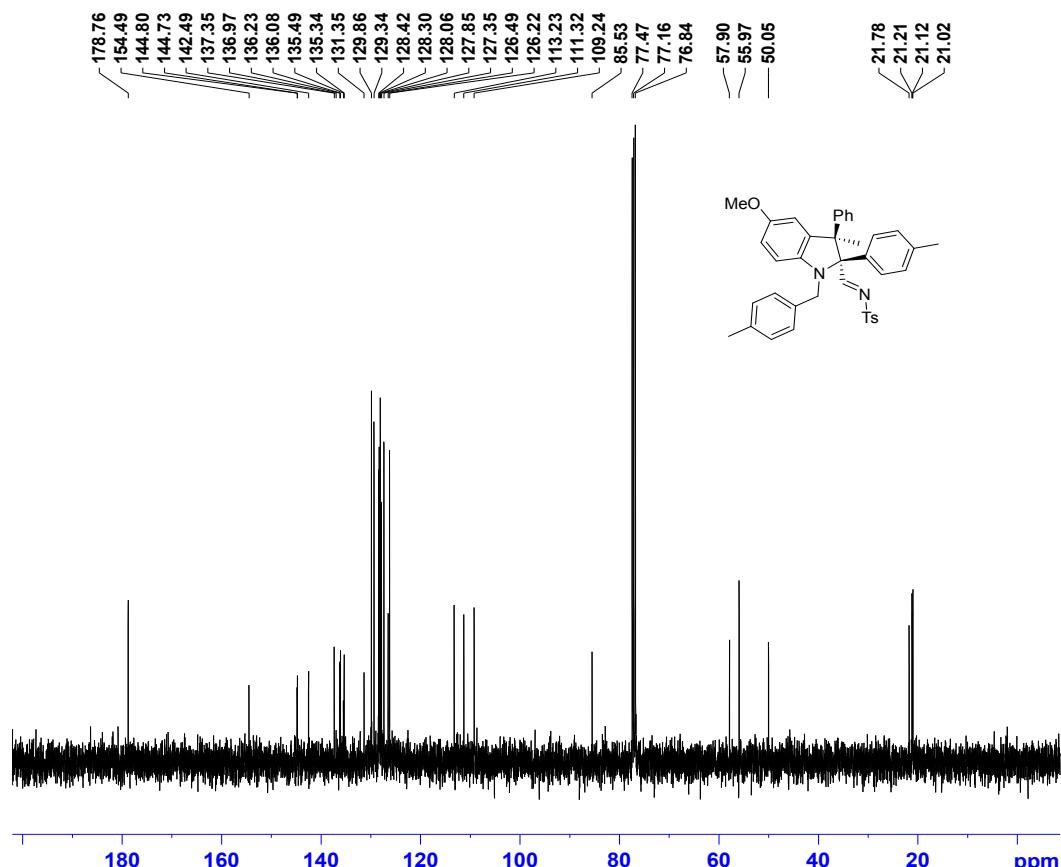
F2 - Processing parameters  
SI 32768  
SF 100.6127544 MHz  
WDW EM  
SSB 0  
LB 1.00 Hz  
GB 0  
PC 1.40

**Indoline (3fa):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

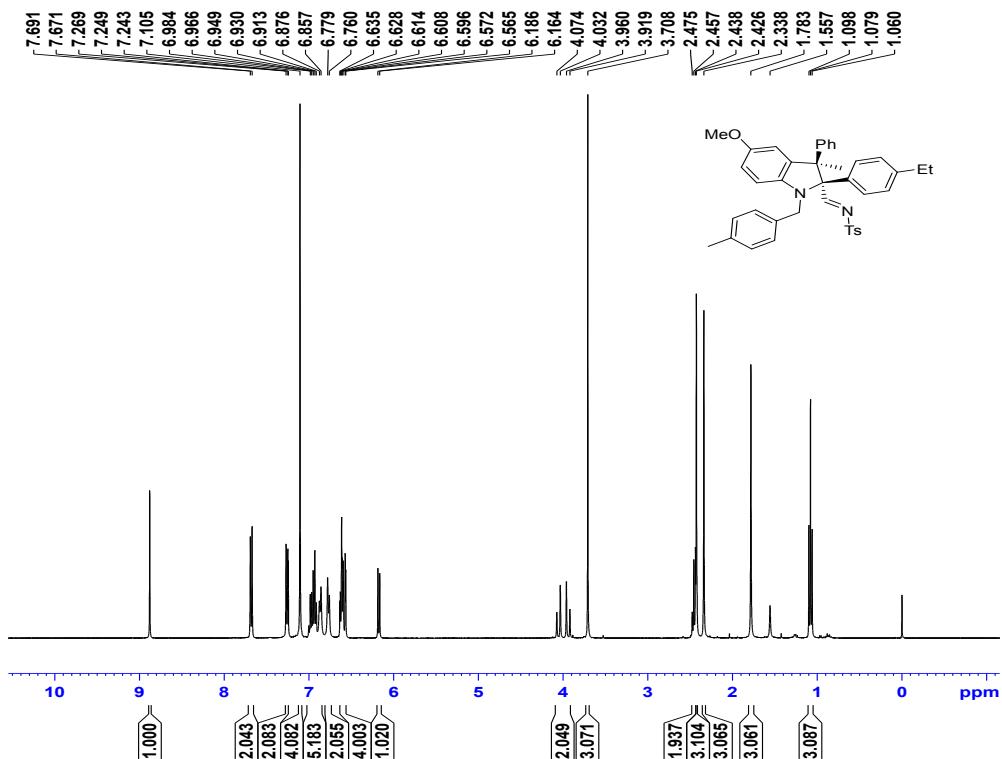


**$^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

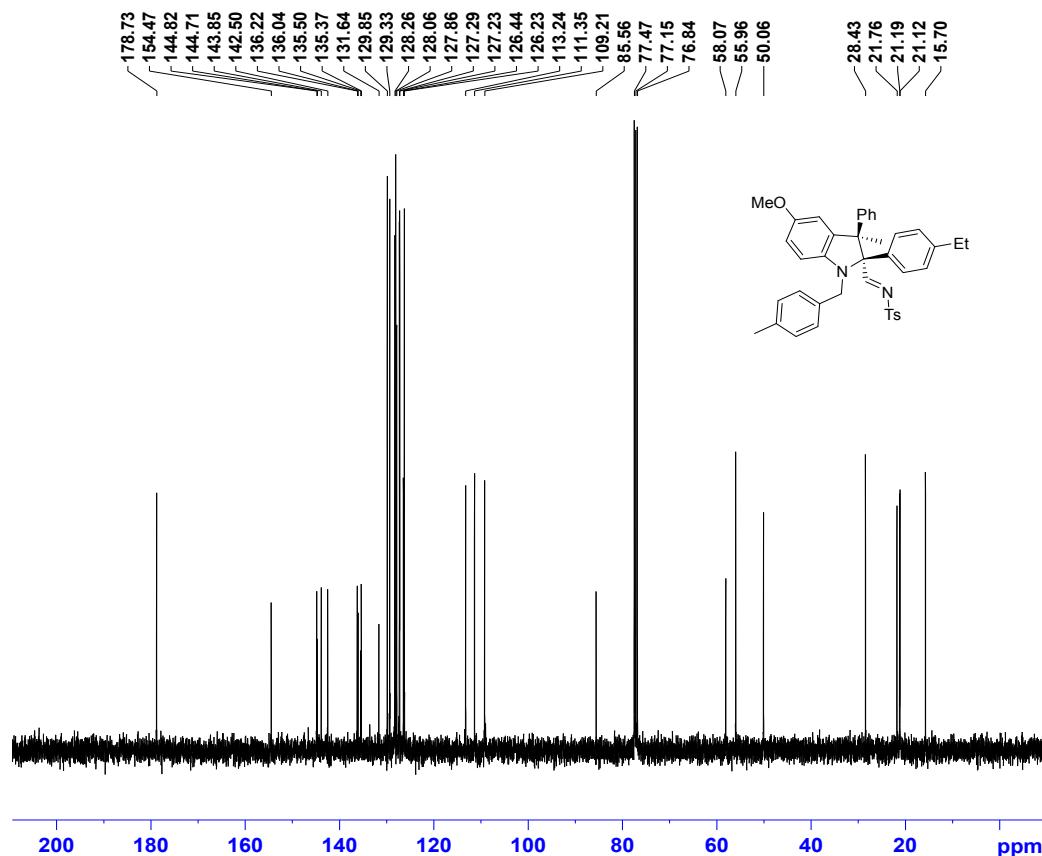


**Indoline (3ga):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

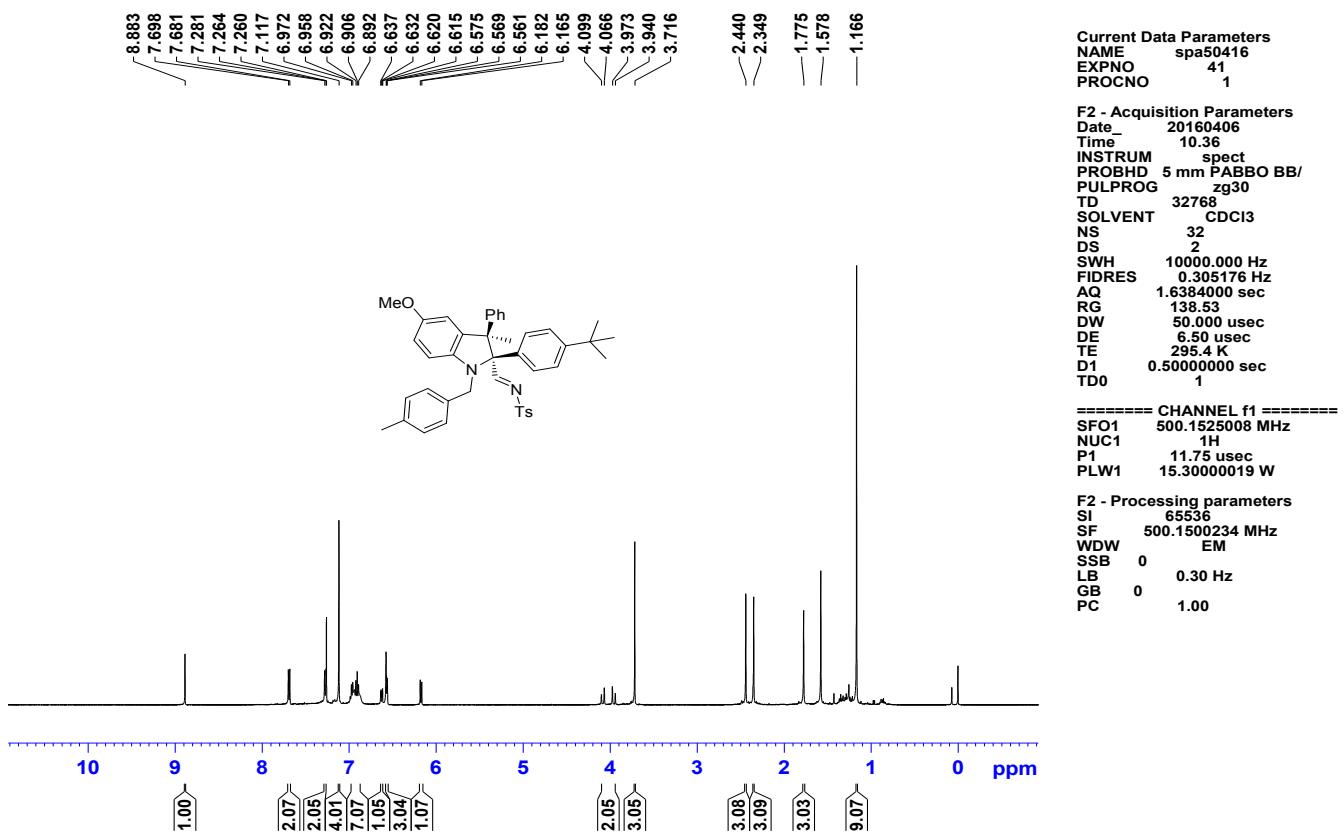


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

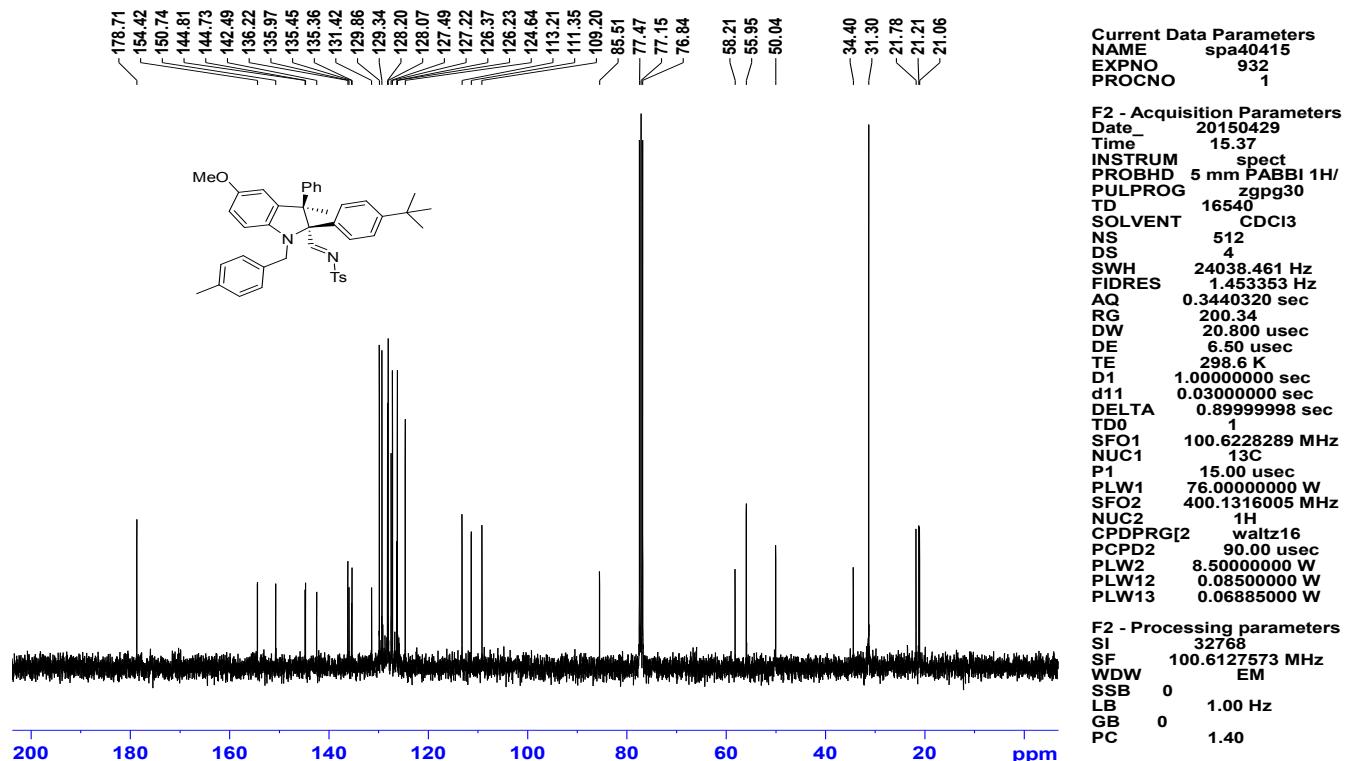


**Indoline (3ha):**

<sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>, 24 °C)

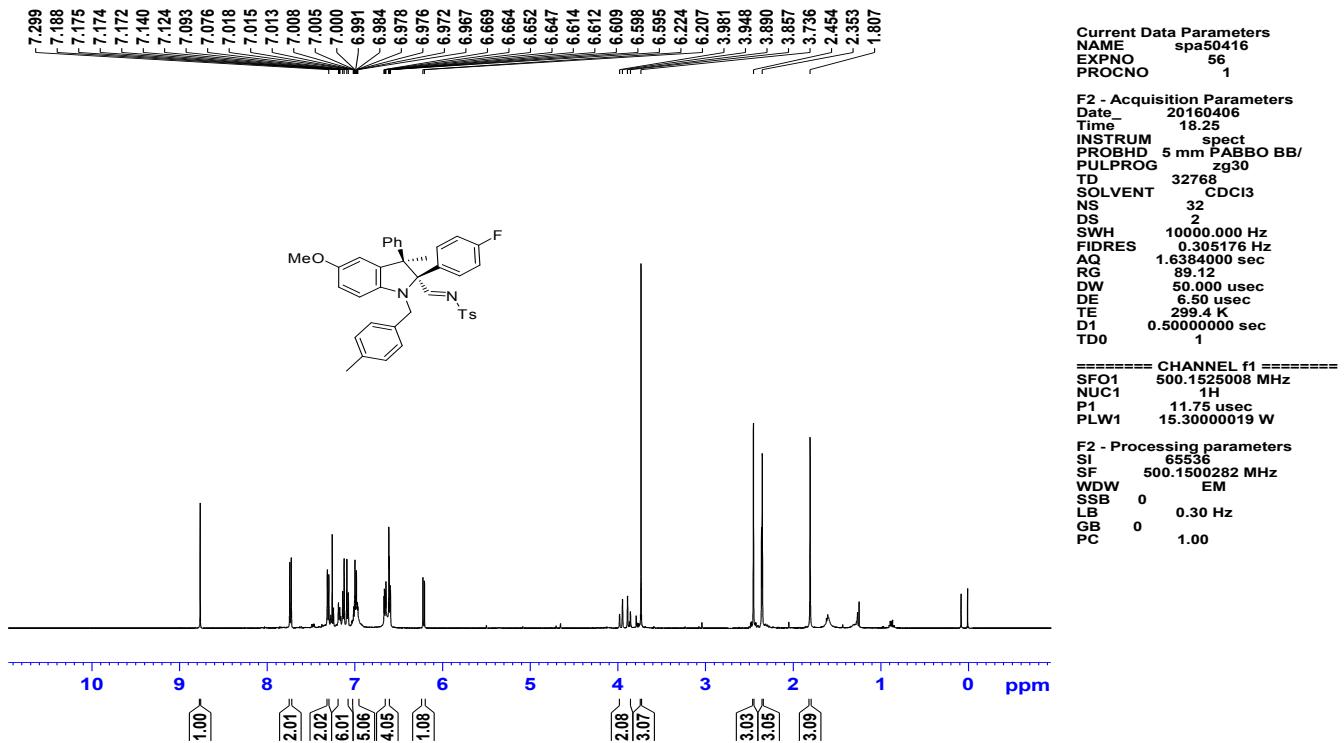


<sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C)

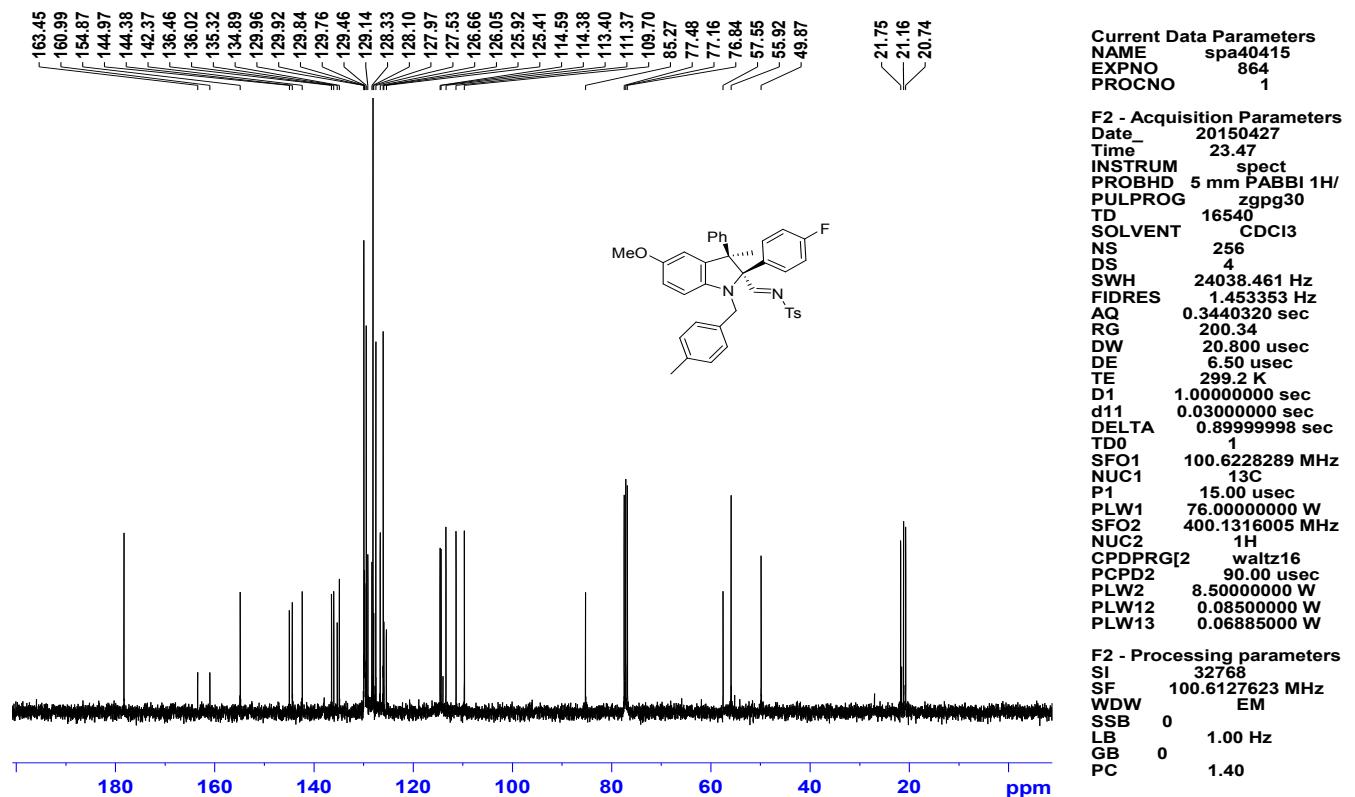


**Indoline (3ia):**

**<sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>, 24 °C)**

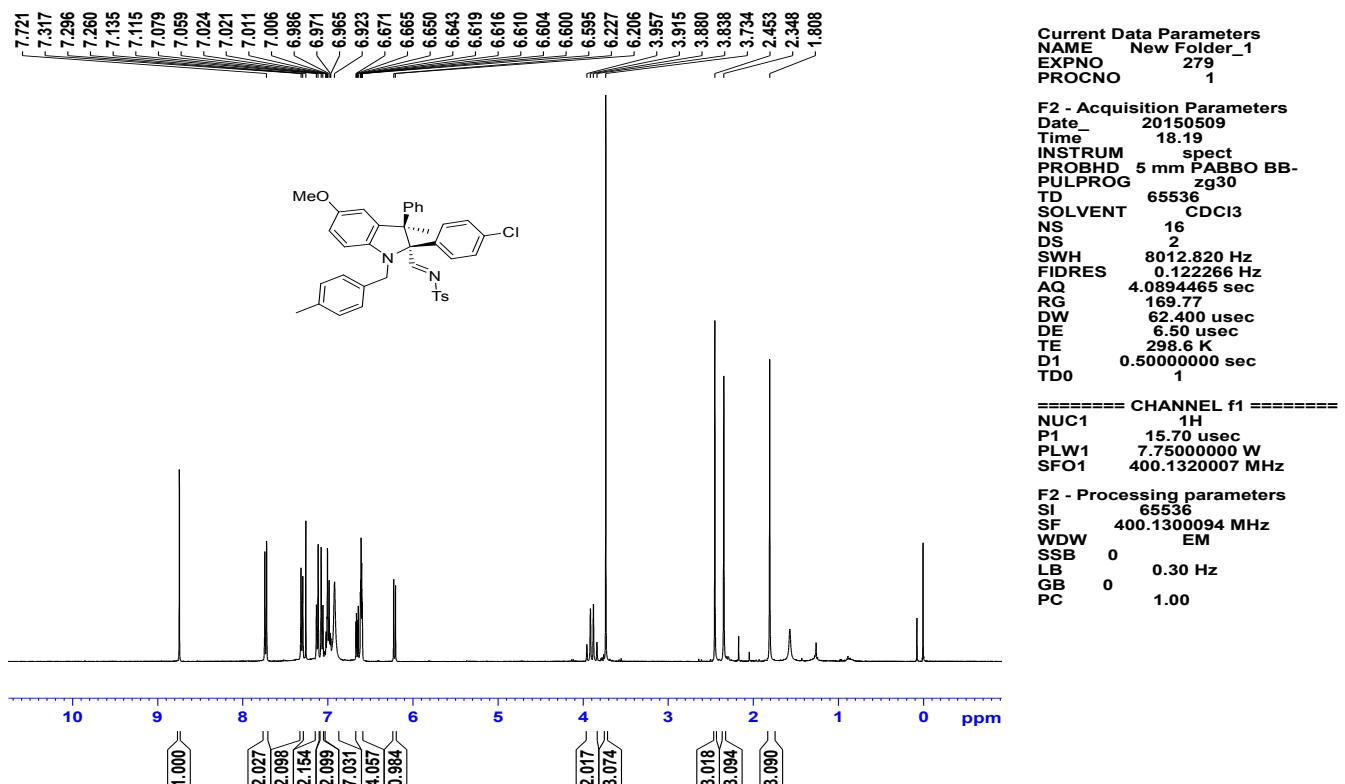


**<sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C)**

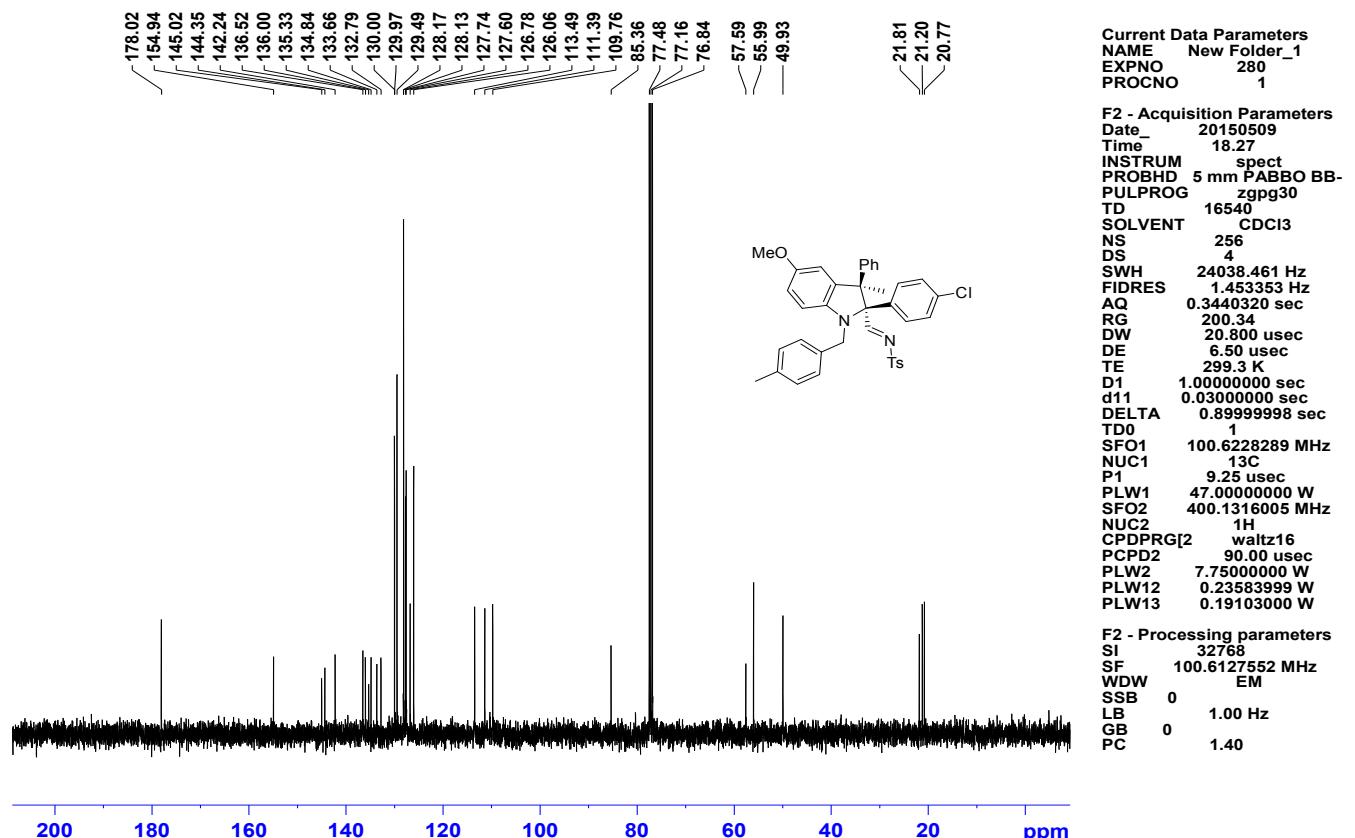


**Indoline (3ja):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

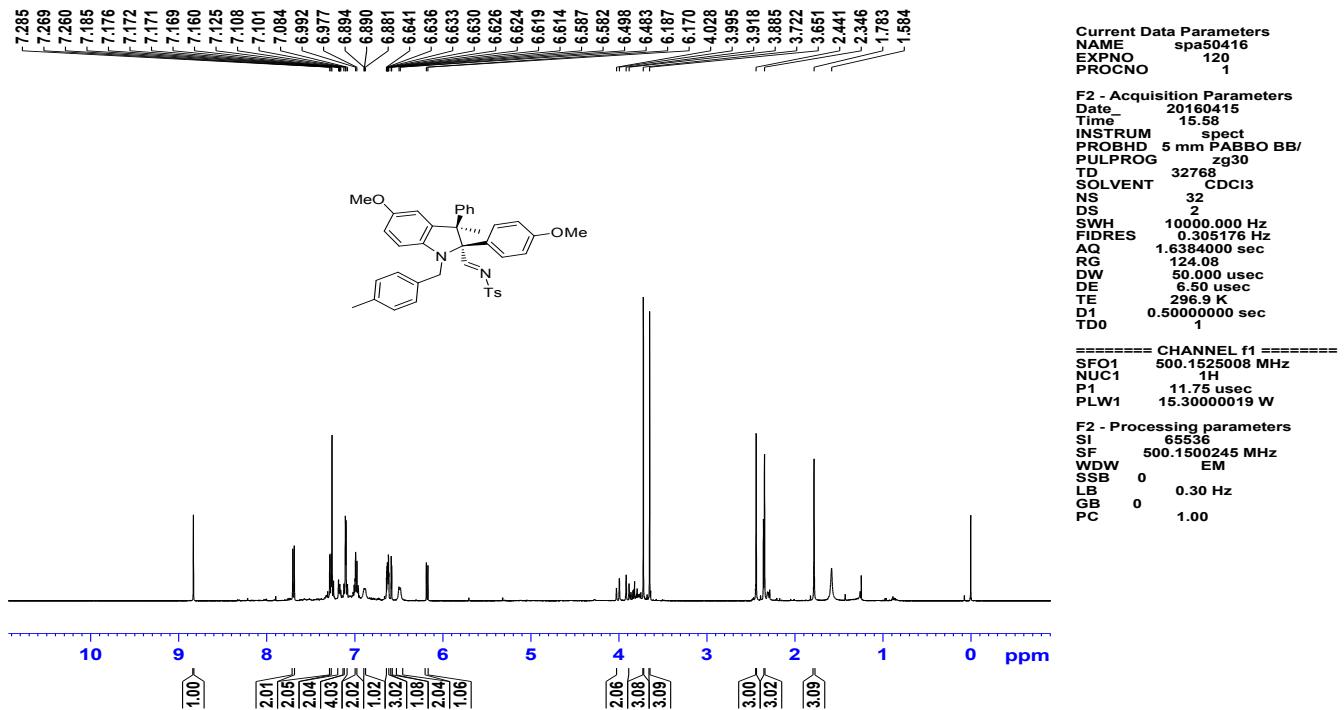


**$^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

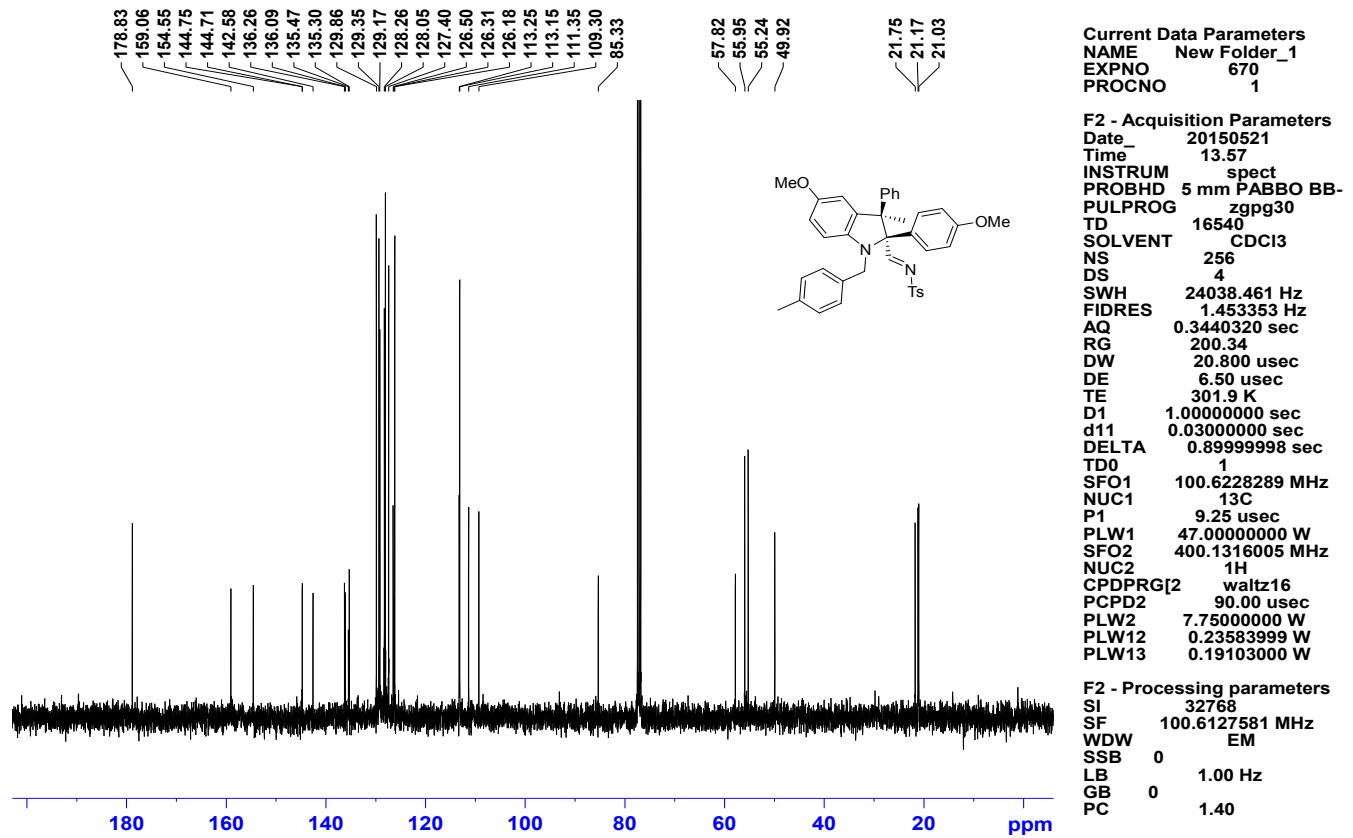


**Indoline (3ka):**

**<sup>1</sup>H NMR (500 MHz, CDCl<sub>3</sub>, 24 °C)**

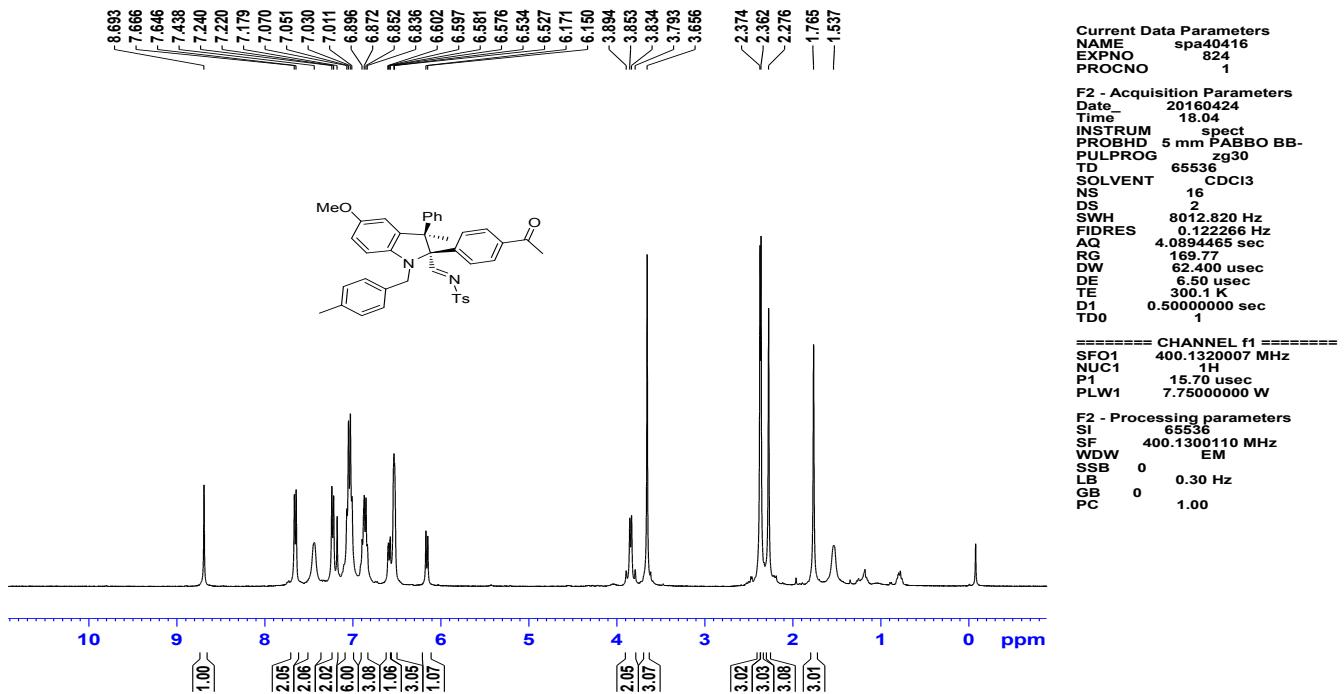


**<sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C)**

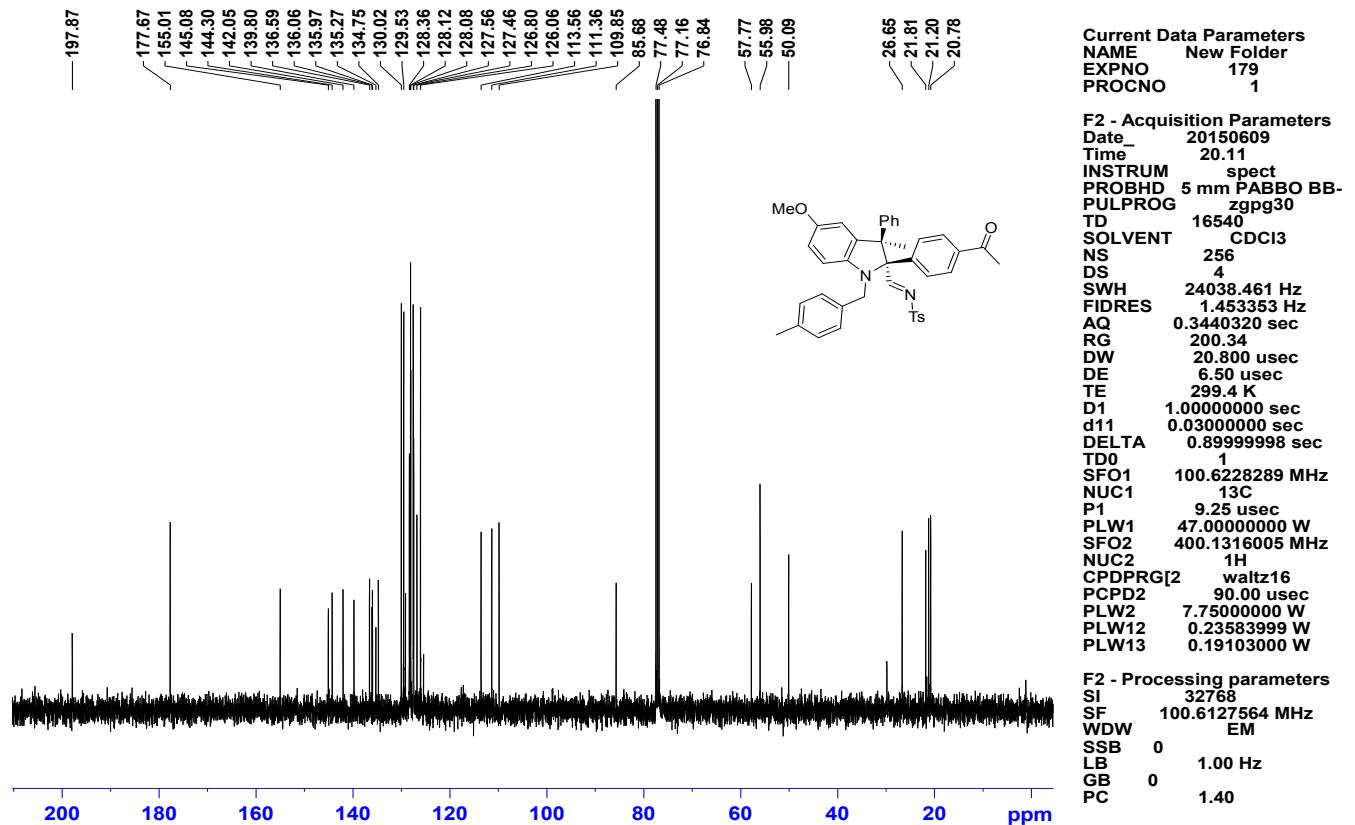


**Indoline (3la):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

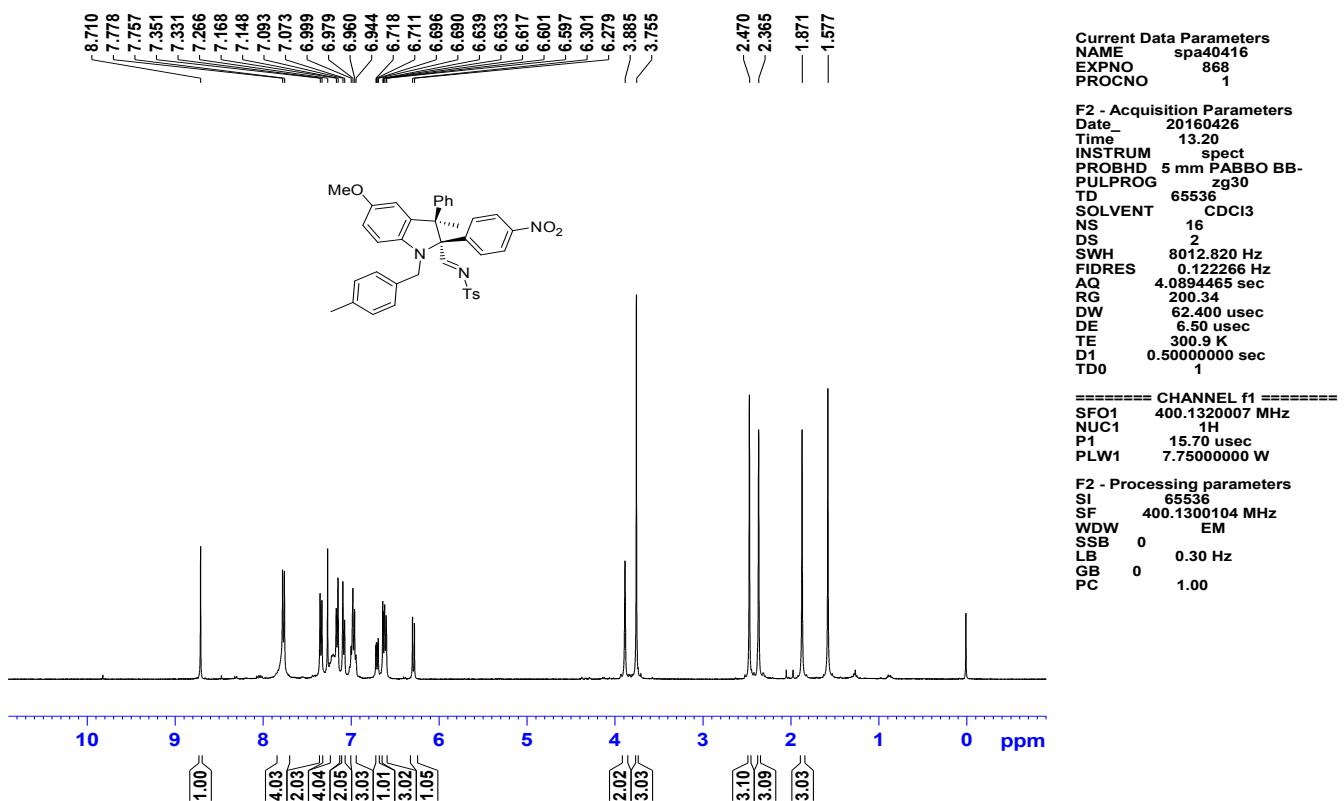


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

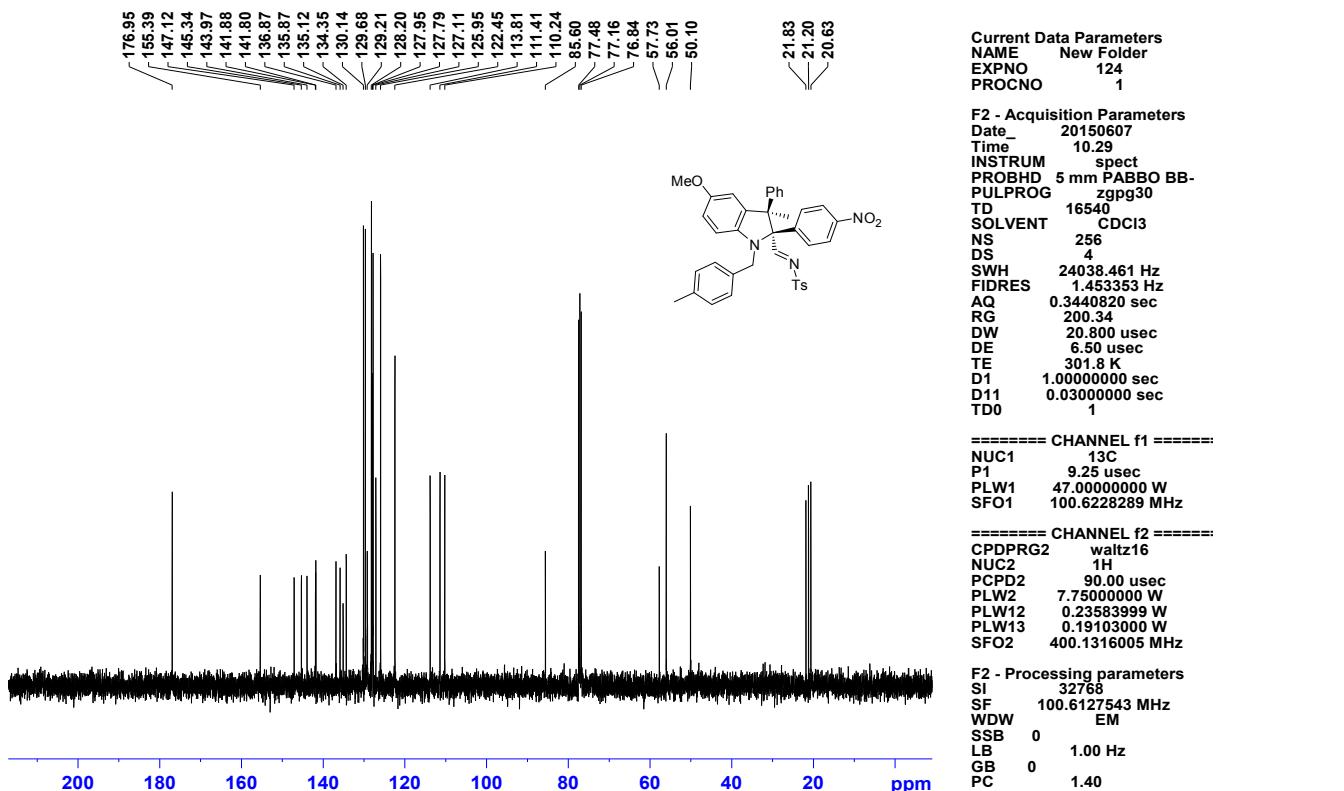


**Indoline (3ma):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

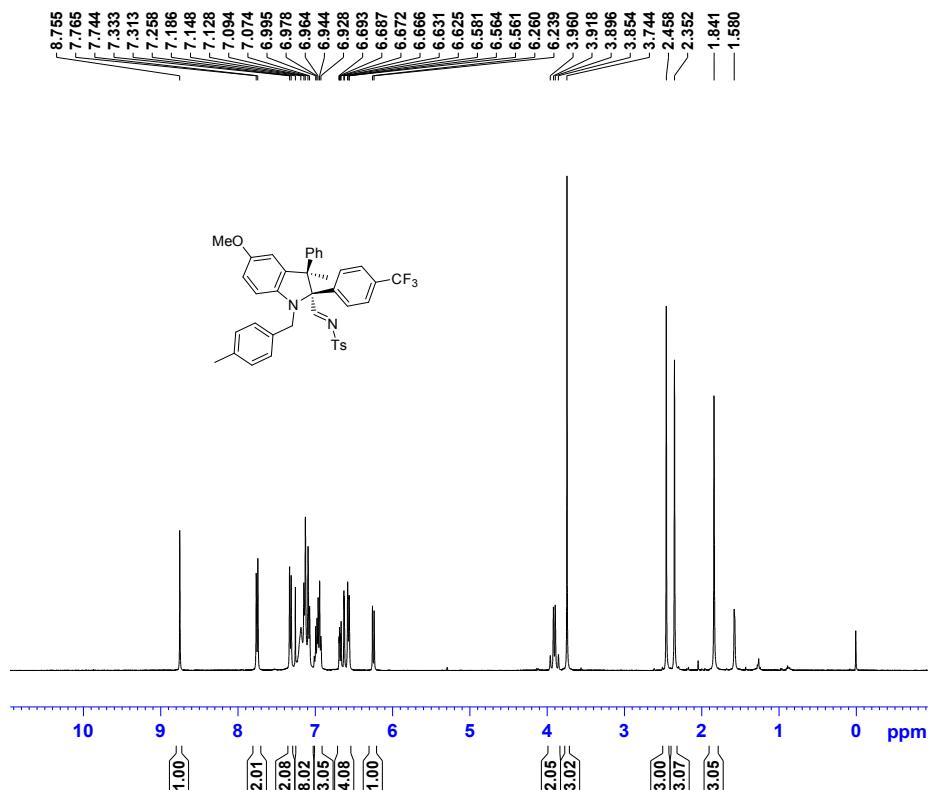


**$^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



**Indoline (3na):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**



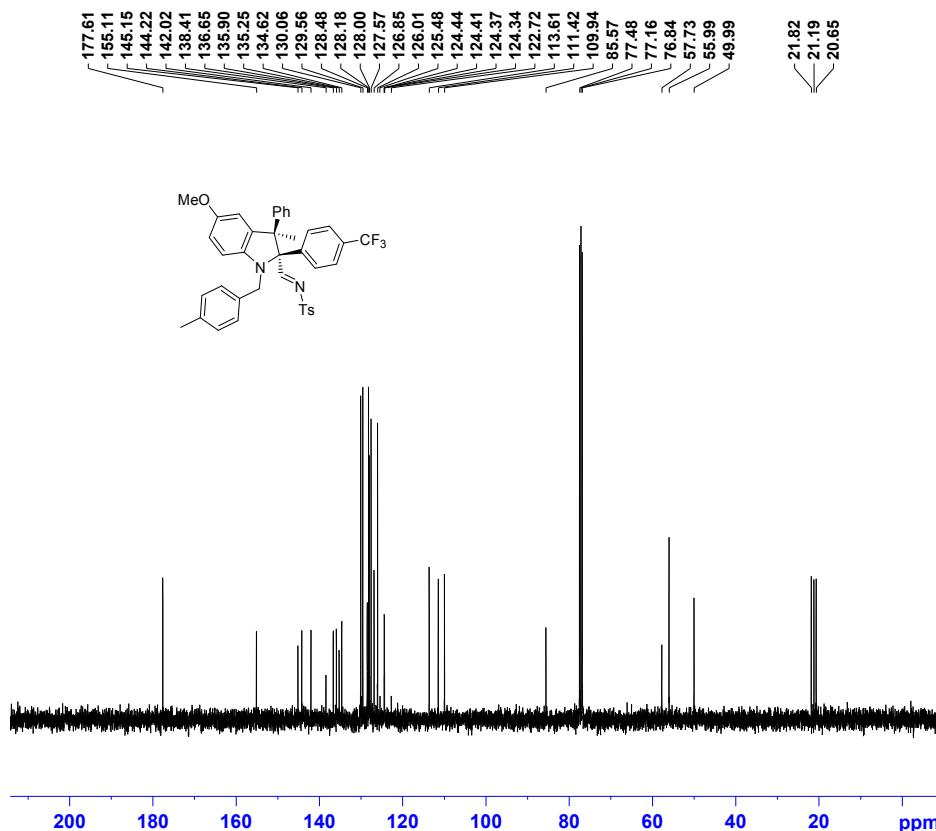
**Current Data Parameters**  
**NAME** New Folder\_1  
**EXPNO** 742  
**PROCNO** 1

**F2 - Acquisition Parameters**  
**Date** 20150524  
**Time** 22.07  
**INSTRUM** spect  
**PROBHD** 5 mm PABBO BB-  
**PULPROG** zg30  
**TD** 65536  
**SOLVENT**  $\text{CDCl}_3$   
**NS** 16  
**DS** 2  
**SWH** 8012.820 Hz  
**FIDRES** 0.122266 Hz  
**AQ** 4.0894966 sec  
**RG** 138.85  
**DW** 62.400 usec  
**DE** 6.50 usec  
**TE** 298.9 K  
**D1** 0.5000000 sec  
**TD0** 1

**===== CHANNEL f1 =====**  
**NUC1**  $^1\text{H}$   
**P1** 15.70 usec  
**PLW1** 7.7500000 W  
**SFO1** 400.1320007 MHz

**F2 - Processing parameters**  
**SI** 65536  
**SF** 400.1300102 MHz  
**WDW** EM  
**SSB** 0  
**LB** 0.30 Hz  
**GB** 0  
**PC** 1.00

**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



**Current Data Parameters**  
**NAME** New Folder\_1  
**EXPNO** 743  
**PROCNO** 1

**F2 - Acquisition Parameters**  
**Date** 20150524  
**Time** 22.10  
**INSTRUM** spect  
**PROBHD** 5 mm PABBO BB-  
**PULPROG** zgpg30  
**TD** 16540  
**SOLVENT**  $\text{CDCl}_3$   
**NS** 256  
**DS** 4  
**SWH** 24038.461 Hz  
**FIDRES** 1.453353 Hz  
**AQ** 0.3440820 sec  
**RG** 200.34  
**DW** 20.800 usec  
**DE** 6.50 usec  
**TE** 299.2 K  
**D1** 1.0000000 sec  
**D11** 0.03000000 sec  
**TD0** 1

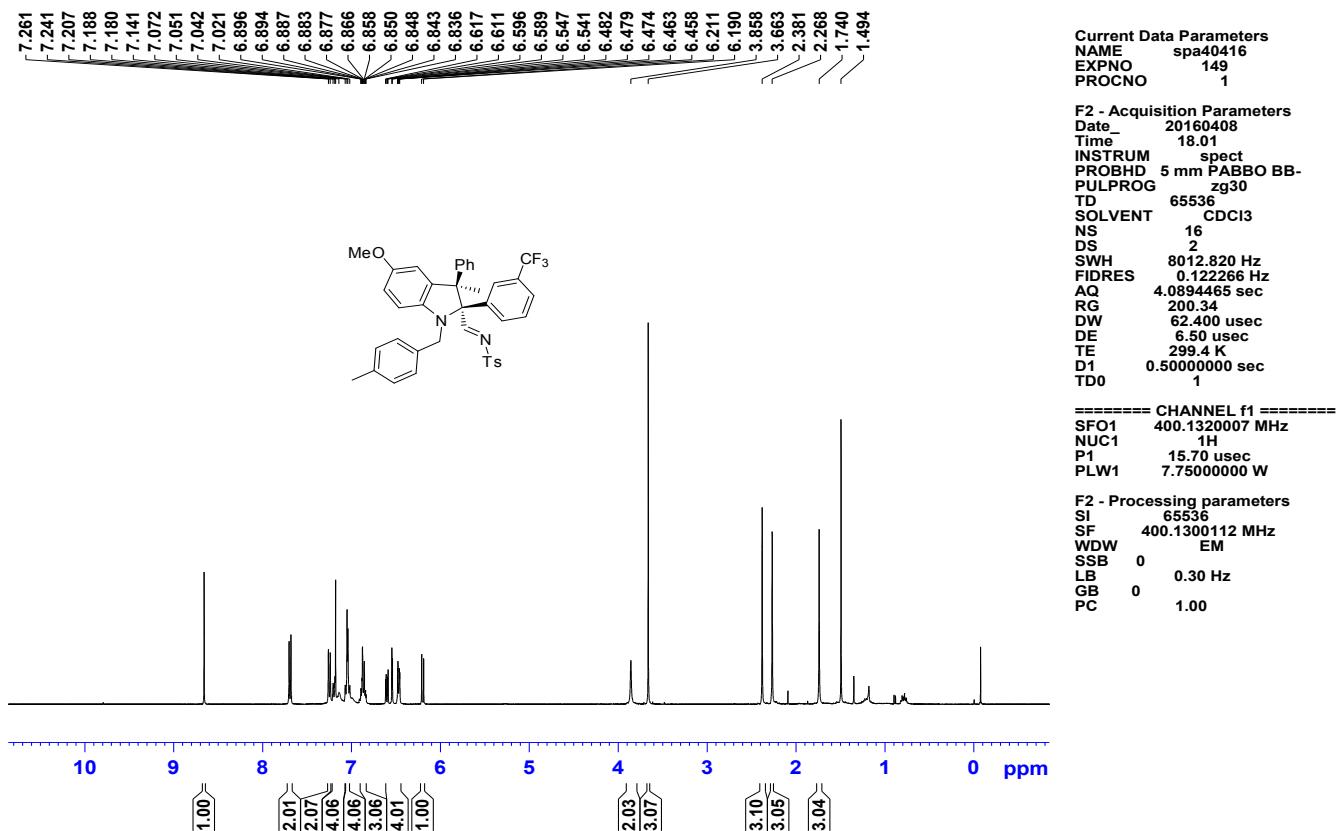
**===== CHANNEL f1 =====**  
**NUC1**  $^{13}\text{C}$   
**P1** 9.25 usec  
**PLW1** 47.00000000 W  
**SFO1** 100.6228289 MHz

**===== CHANNEL f2 =====**  
**CPDPRG2** waltz16  
**NUC2**  $^1\text{H}$   
**PCPD2** 90.00 usec  
**PLW2** 7.75000000 W  
**PLW12** 0.23583999 W  
**PLW13** 0.19103000 W  
**SFO2** 400.1316005 MHz

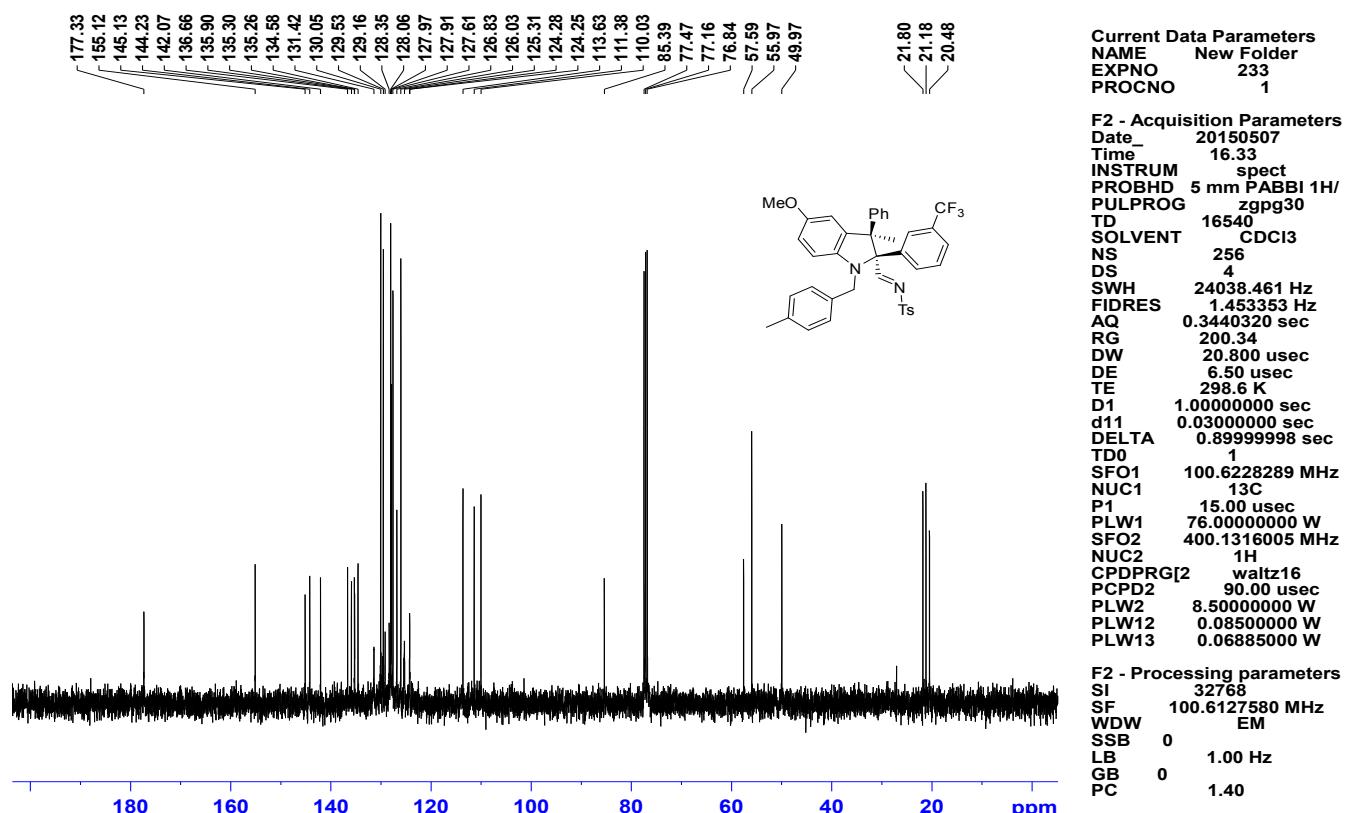
**F2 - Processing parameters**  
**SI** 32768  
**SF** 100.6127553 MHz  
**WDW** EM  
**SSB** 0  
**LB** 1.00 Hz  
**GB** 0  
**PC** 1.40

**Indoline (3oa):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

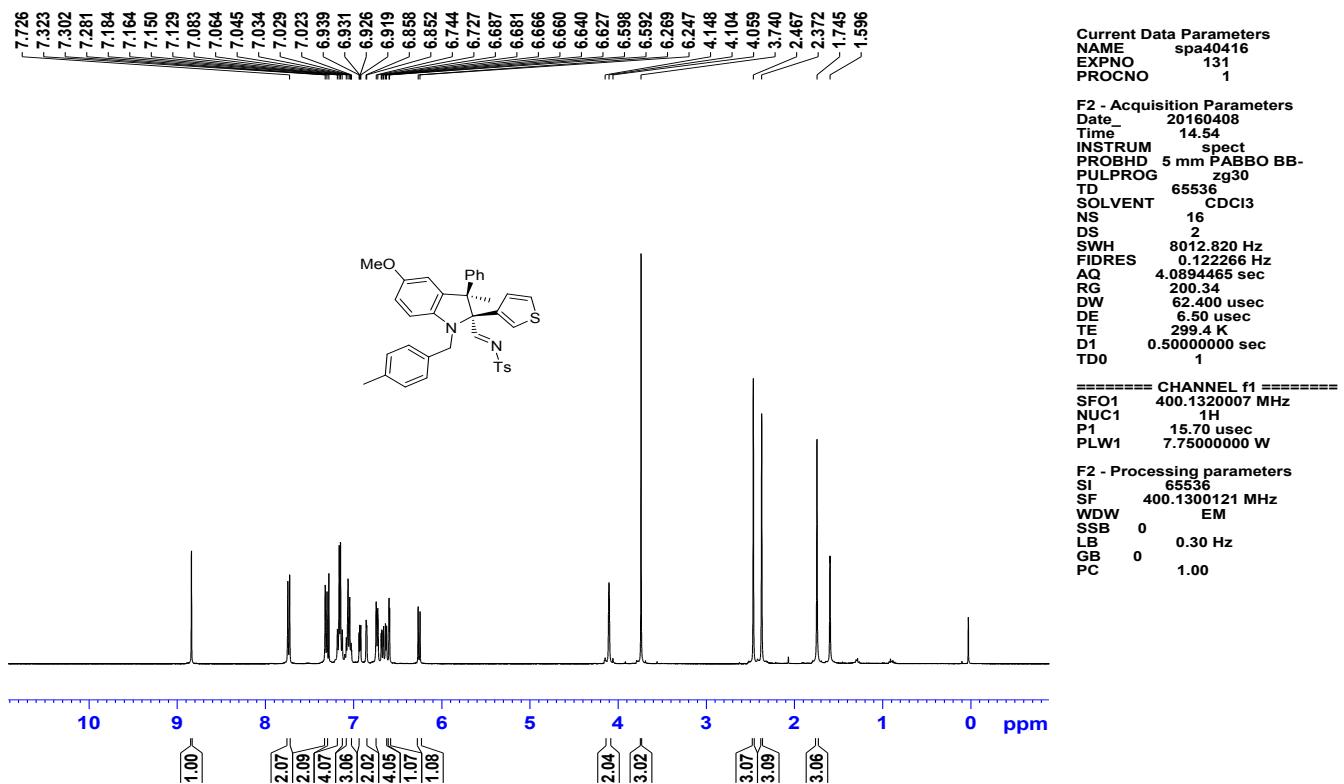


**$^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

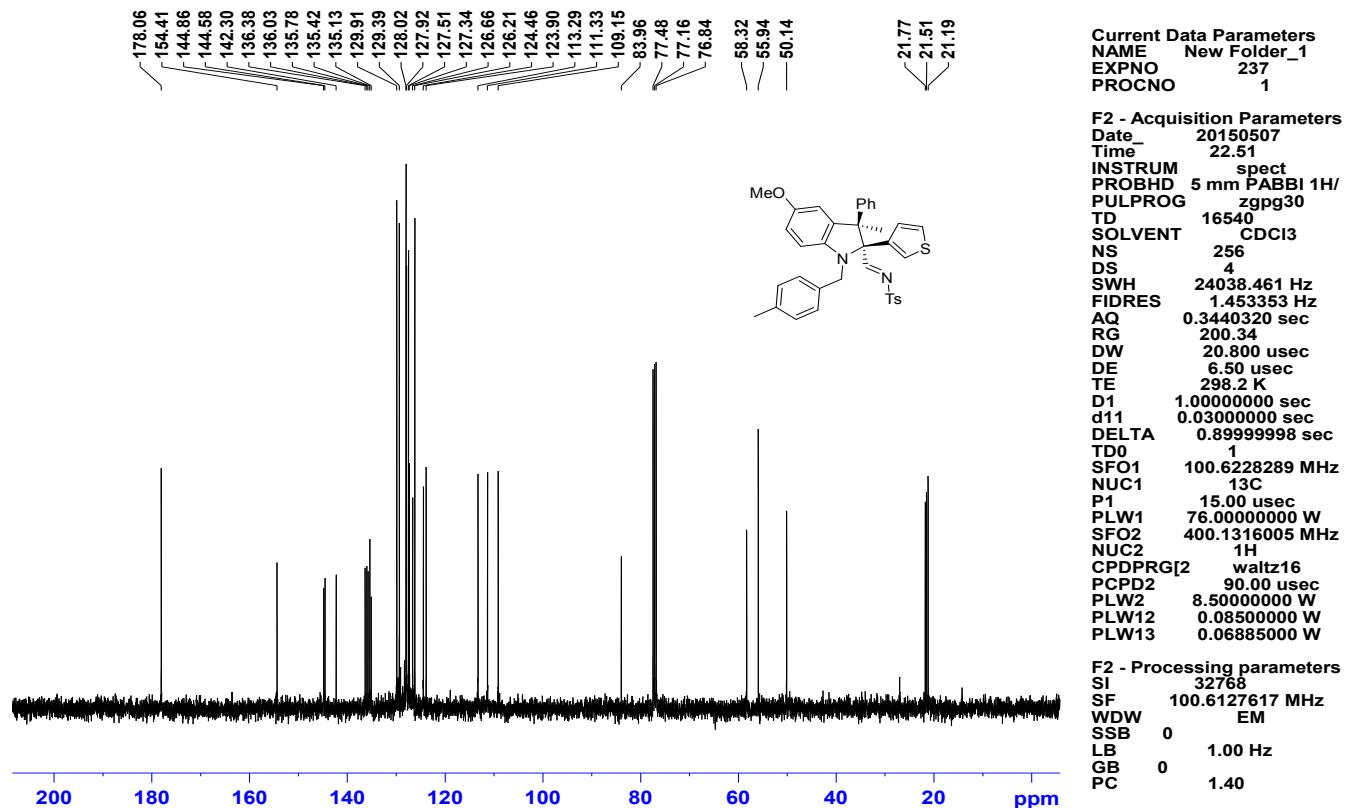


**Indoline (3pa):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

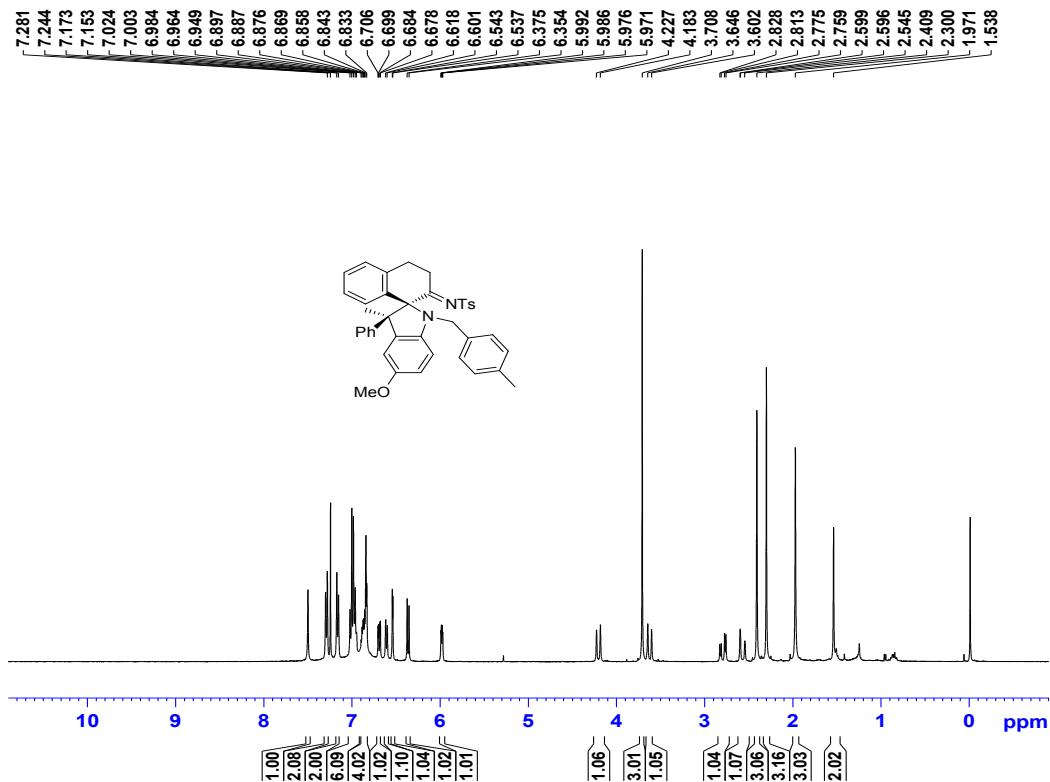


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



**Indoline (3qa):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**



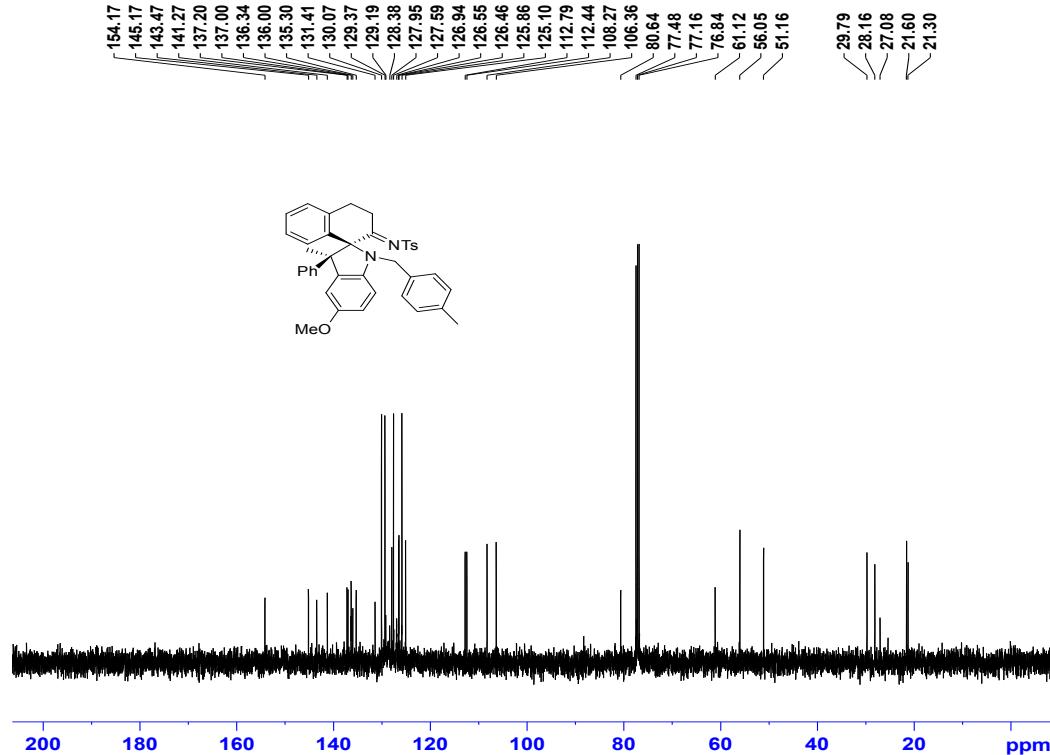
Current Data Parameters  
 NAME spa40416  
 EXPNO 614  
 PROCNO 1

F2 - Acquisition Parameters  
 Date\_ 20160420  
 Time 2.15  
 INSTRUM spect  
 PROBHD 5 mm PABBO BB-  
 PULPROG zg30  
 TD 65536  
 SOLVENT CDCl3  
 NS 16  
 DS 2  
 SWH 8012.80 Hz  
 FIDRES 0.122266 Hz  
 AQ 4.0894465 sec  
 RG 200.34  
 DW 62.400 usec  
 DE 6.50 usec  
 TE 299.9 K  
 D1 0.5000000 sec  
 TD0 1

===== CHANNEL f1 ======  
 SFO1 400.1320007 MHz  
 NUC1 1H  
 P1 15.70 usec  
 PLW1 7.7500000 W

F2 - Processing parameters  
 SI 65536  
 SF 400.1300113 MHz  
 WDW EM  
 SSB 0  
 LB 0.30 Hz  
 GB 0  
 PC 1.00

**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



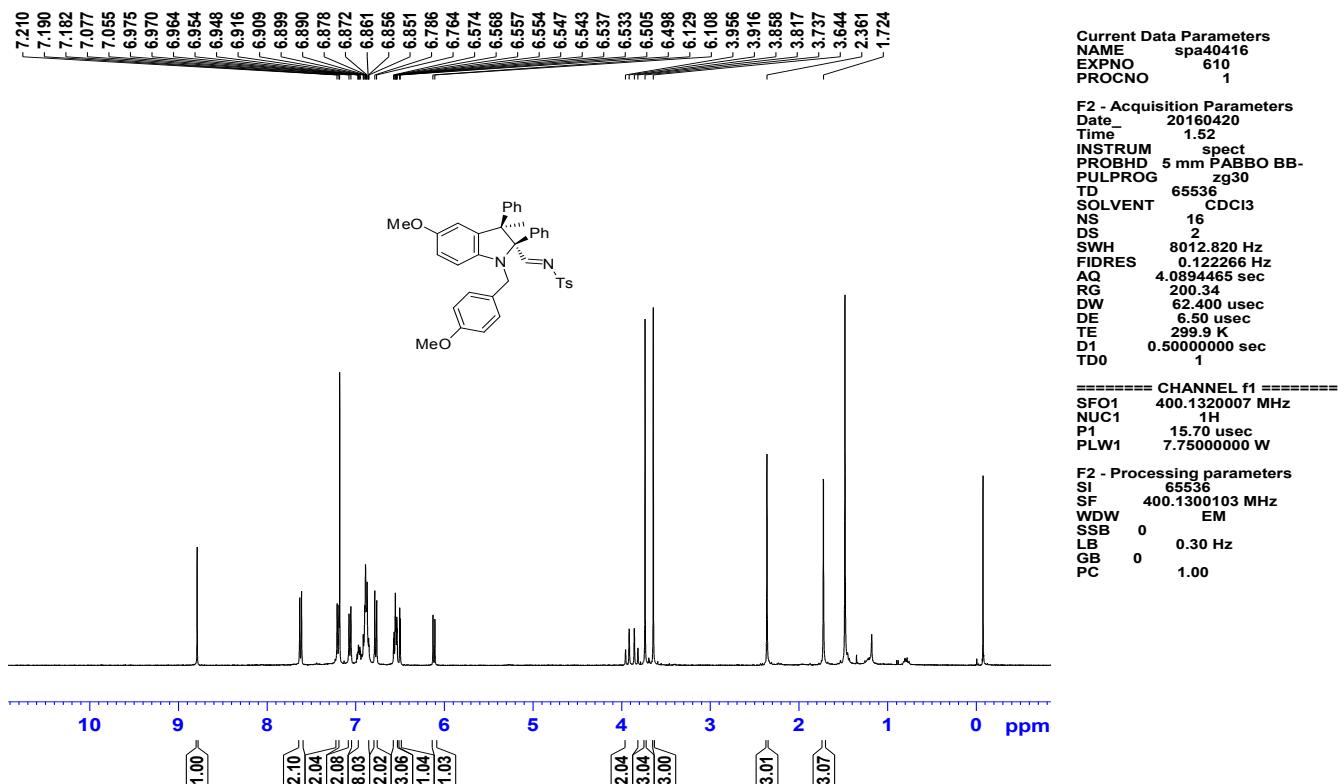
Current Data Parameters  
 NAME New Folder  
 EXPNO 126  
 PROCNO 1

F2 - Acquisition Parameters  
 Date\_ 20150607  
 Time 10.43  
 INSTRUM spect  
 PROBHD 5 mm PABBO BB-  
 PULPROG zgpg30  
 TD 16540  
 SOLVENT CDCl3  
 NS 256  
 DS 4  
 SWH 24038.461 Hz  
 FIDRES 1.45353 Hz  
 AQ 0.3440320 sec  
 RG 200.34  
 DW 20.800 usec  
 DE 6.50 usec  
 TE 301.7 K  
 D1 1.0000000 sec  
 d11 0.0300000 sec  
 DELTA 0.8999999 sec  
 TD0 1  
 SFO1 100.6228289 MHz  
 NUC1 13C  
 P1 9.25 usec  
 PLW1 47.0000000 W  
 SFO2 400.1316005 MHz  
 NUC2 1H  
 CPDPRG[2] waltz16  
 PCPD2 90.00 usec  
 PLW2 7.7500000 W  
 PLW12 0.23583999 W  
 PLW13 0.19103000 W

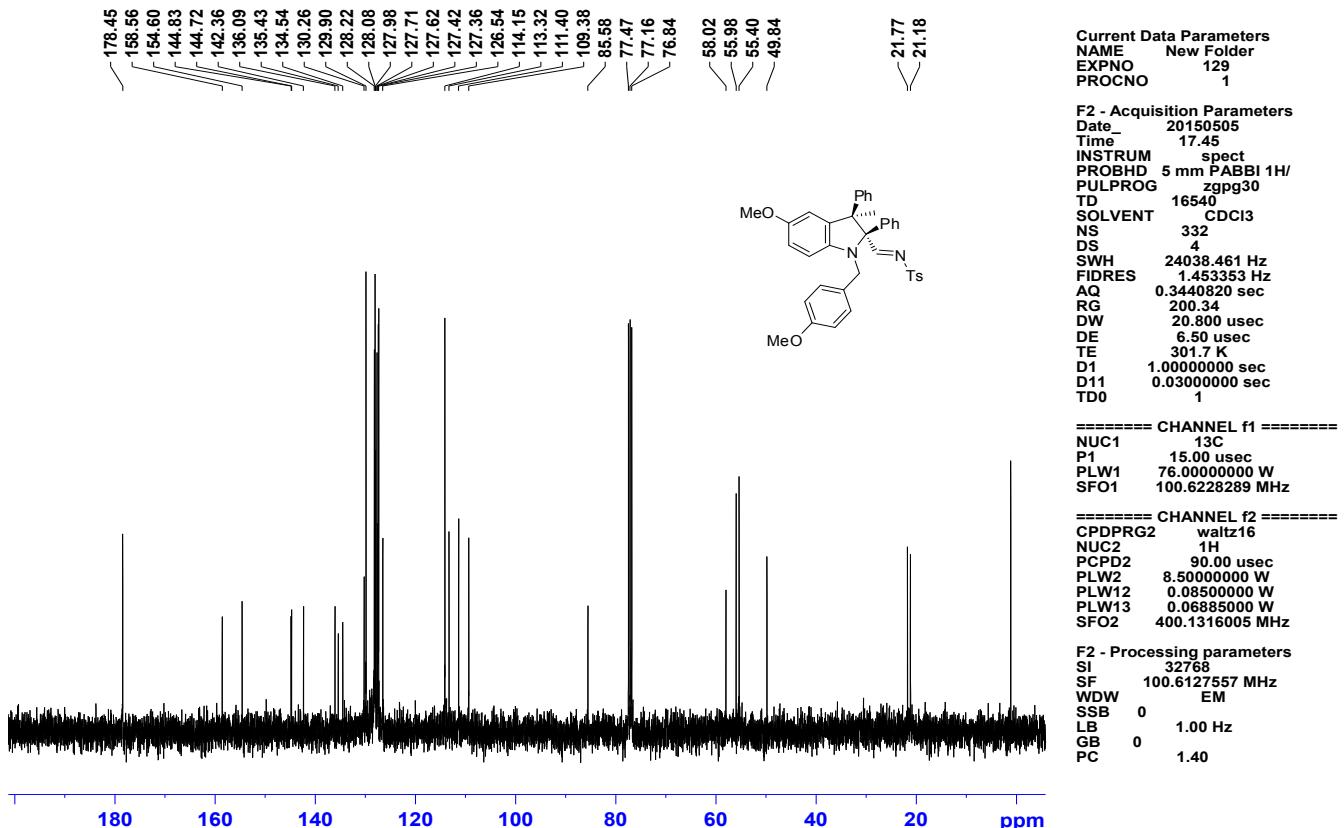
F2 - Processing parameters  
 SI 32768  
 SF 100.6127527 MHz  
 WDW EM  
 SSB 0  
 LB 1.00 Hz  
 GB 0  
 PC 1.40

**Indoline (3ab):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

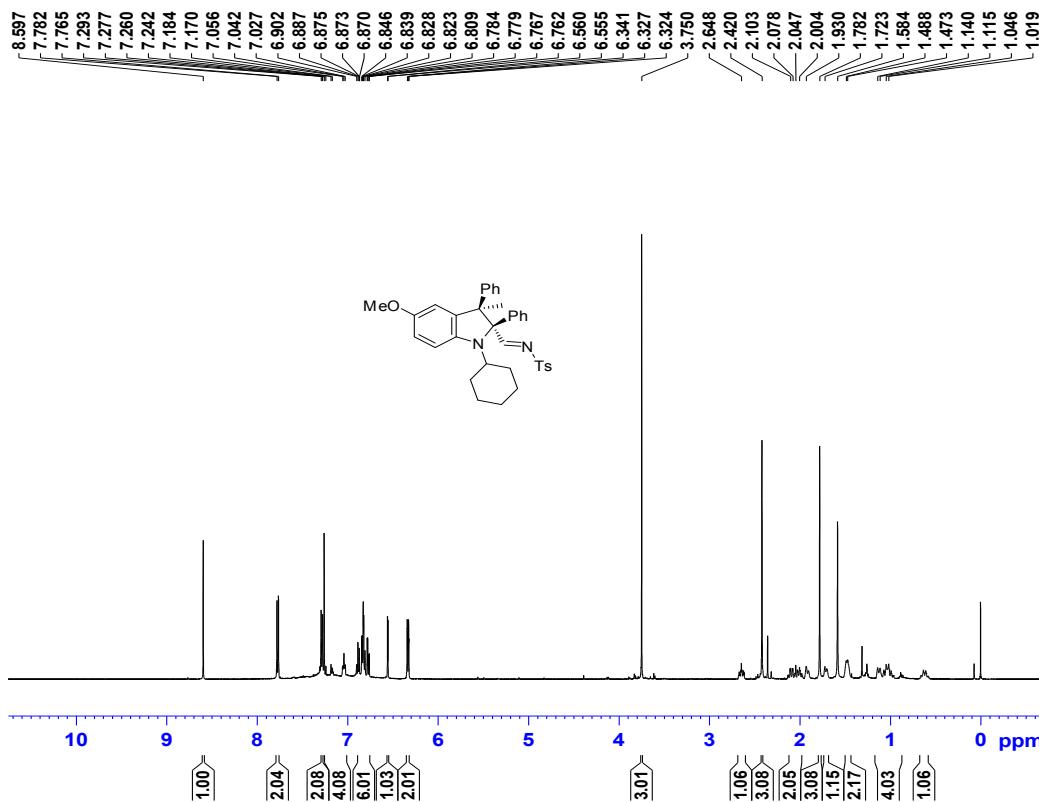


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



**Indoline (3ac):**

**$^1\text{H}$  NMR (500 MHz,  $\text{CDCl}_3$ , 24 °C)**



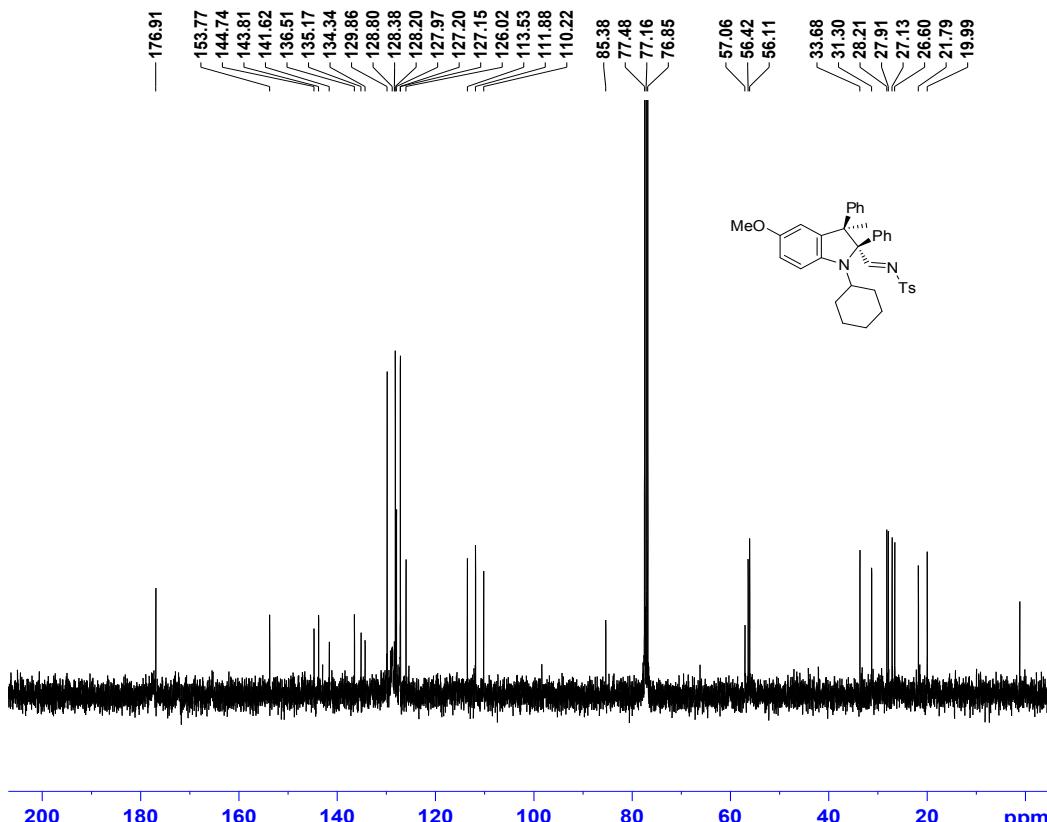
Current Data Parameters  
NAME spa50416  
EXPNO 80  
PROCNO 1

F2 - Acquisition Parameters  
Date 20160407  
Time 14.24  
INSTRUM spect  
PROBHD 5 mm PABBO BB/  
PULPROG zg30  
TD 32768  
SOLVENT CDCl3  
NS 32  
DS 2  
SWH 10000.000 Hz  
FIDRES 0.305176 Hz  
AQ 1.6384000 sec  
RG 114.76  
DW 50.000 usec  
DE 6.50 usec  
TE 297.8 K  
D1 0.5000000 sec  
TD0 1

===== CHANNEL f1 ======  
SFO1 500.1525008 MHz  
NUC1 1H  
P1 11.75 usec  
PLW1 15.30000019 W

F2 - Processing parameters  
SI 65536  
SF 500.1500242 MHz  
WDW EM  
SSB 0  
LB 0.30 Hz  
GB 0  
PC 1.00

**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



Current Data Parameters  
NAME New Folder  
EXPNO 252  
PROCNO 1

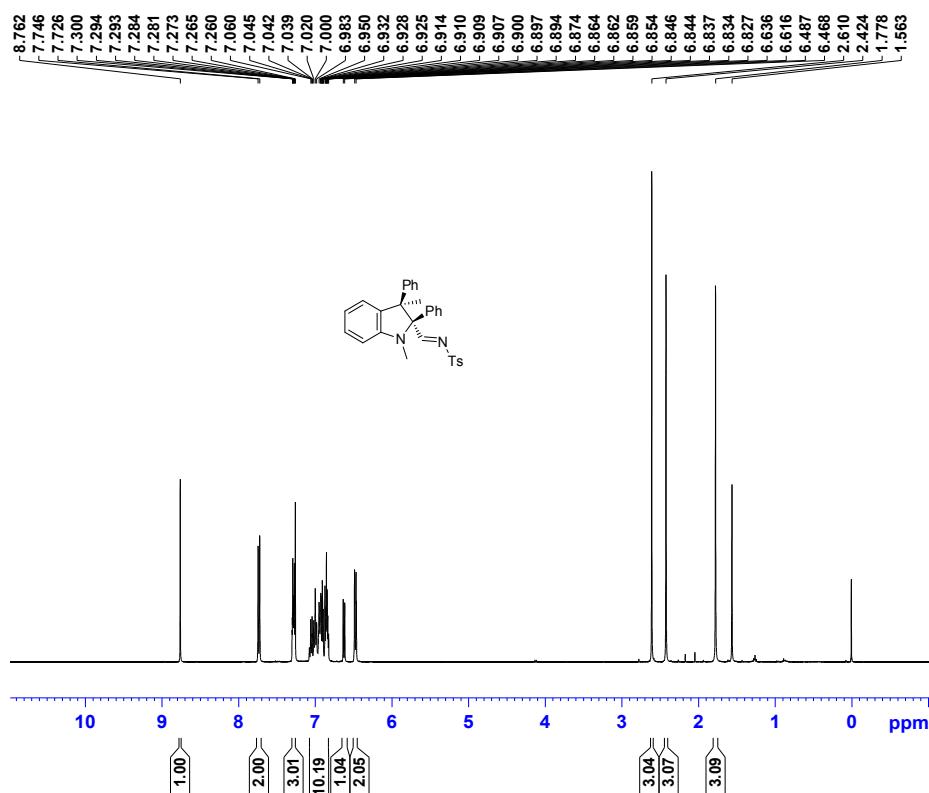
F2 - Acquisition Parameters  
Date 20150508  
Time 4.45  
INSTRUM spect  
PROBHD 5 mm PABBI 1H/  
PULPROG zgpg30  
TD 16540  
SOLVENT CDCl3  
NS 2000  
DS 4  
SWH 24038.461 Hz  
FIDRES 1.453353 Hz  
AQ 0.3440820 sec  
RG 200.34  
DW 20.800 usec  
DE 6.50 usec  
TE 298.4 K  
D1 1.0000000 sec  
D11 0.0300000 sec  
TD0 1

===== CHANNEL f1 ======  
NUC1 13C  
P1 15.00 usec  
PLW1 76.0000000 W  
SFO1 100.6228289 MHz

===== CHANNEL f2 ======  
CPDPRG2 waltz16  
NUC2 1H  
PCPD2 90.00 usec  
PLW2 8.5000000 W  
PLW12 0.0850000 W  
PLW13 0.06885000 W  
SFO2 400.1316005 MHz  
F2 - Processing parameters  
SI 32768  
SF 100.6127544 MHz  
WDW EM  
SSB 0  
LB 1.00 Hz  
GB 0  
PC 1.40

**Indoline (3ad):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**



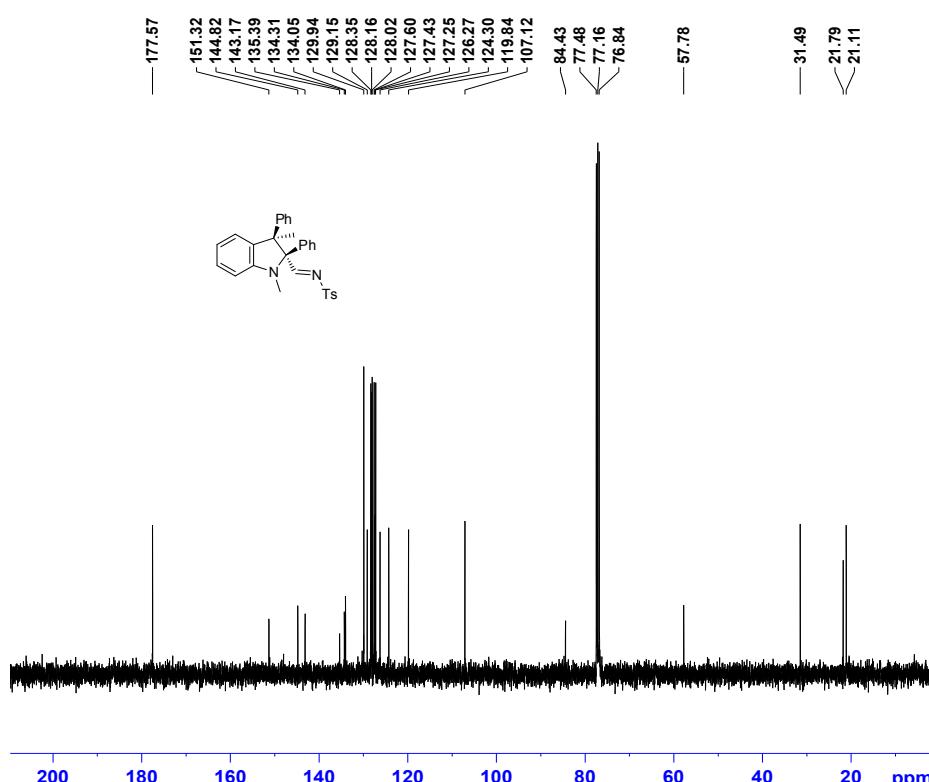
Current Data Parameters  
 NAME New Folder  
 EXPNO 101  
 PROCNO 1

F2 - Acquisition Parameters  
 Date 20150504  
 Time 20.24  
 INSTRUM spect  
 PROBHD 5 mm PABBI 1H/  
 PULPROG zg30  
 TD 65536  
 SOLVENT CDCl3  
 NS 16  
 DS 2  
 SWH 8012.820 Hz  
 FIDRES 0.122266 Hz  
 AQ 4.0894966 sec  
 RG 124.58  
 DW 62.400 usec  
 DE 6.50 usec  
 TE 298.4 K  
 D1 0.5000000 sec  
 TDO 1

===== CHANNEL f1 ======  
 NUC1 1H  
 P1 9.00 usec  
 PLW1 8.5000000 W  
 SFO1 400.1320007 MHz

F2 - Processing parameters  
 SI 65536  
 SF 400.1300102 MHz  
 WDW EM  
 SSB 0  
 LB 0.30 Hz  
 GB 0  
 PC 1.00

**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



Current Data Parameters  
 NAME New Folder  
 EXPNO 99  
 PROCNO 1

F2 - Acquisition Parameters  
 Date 20150504  
 Time 20.18  
 INSTRUM spect  
 PROBHD 5 mm PABBI 1H/  
 PULPROG zgpg30  
 TD 16540  
 SOLVENT CDCl3  
 NS 512  
 DS 4  
 SWH 24038.461 Hz  
 FIDRES 1.453353 Hz  
 AQ 0.3440820 sec  
 RG 200.34  
 DW 20.800 usec  
 DE 6.50 usec  
 TE 298.7 K  
 D1 1.0000000 sec  
 D11 0.03000000 sec  
 TDO 1

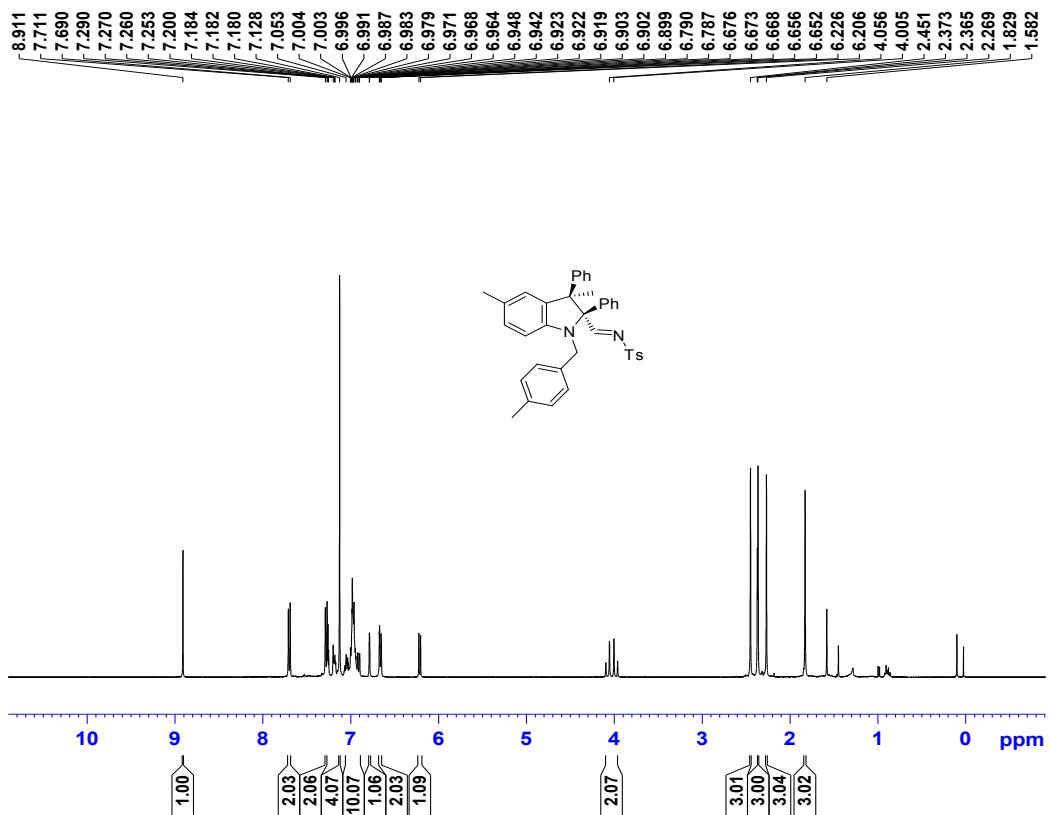
===== CHANNEL f1 ======  
 NUC1 13C  
 P1 15.00 usec  
 PLW1 76.0000000 W  
 SFO1 100.6228289 MHz

===== CHANNEL f2 ======  
 CPDPRG2 waltz16  
 NUC2 1H  
 PCPD2 90.00 usec  
 PLW2 8.5000000 W  
 PLW12 0.08500000 W  
 PLW13 0.06885000 W  
 SFO2 400.1316005 MHz

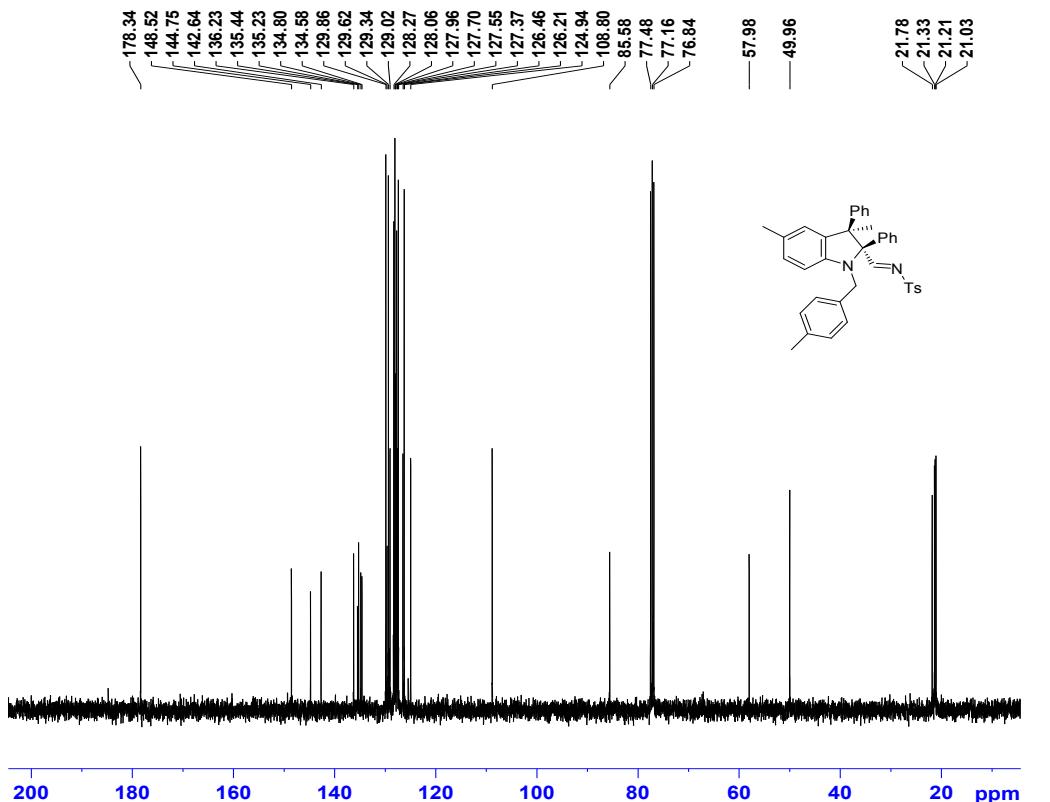
F2 - Processing parameters  
 SI 32768  
 SF 100.6127551 MHz  
 WDW EM  
 SSB 0  
 LB 1.00 Hz  
 GB 0  
 PC 1.40

**Indoline (3af):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

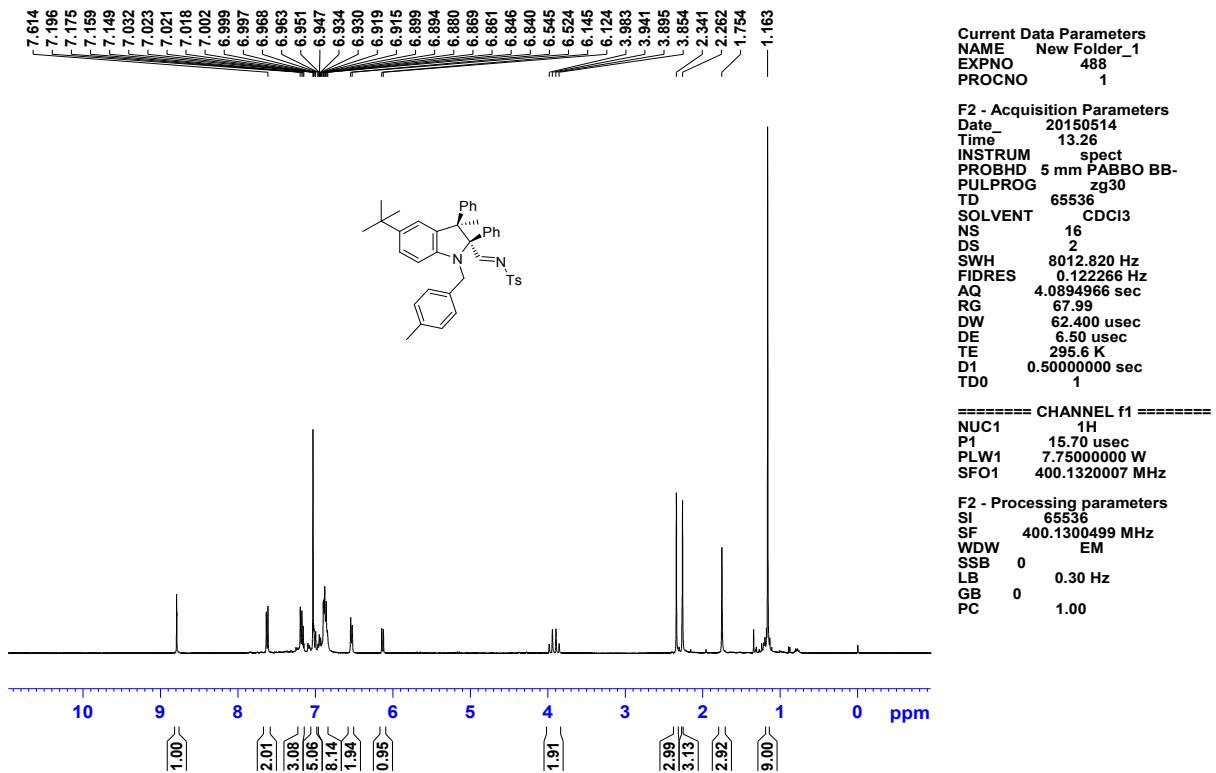


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

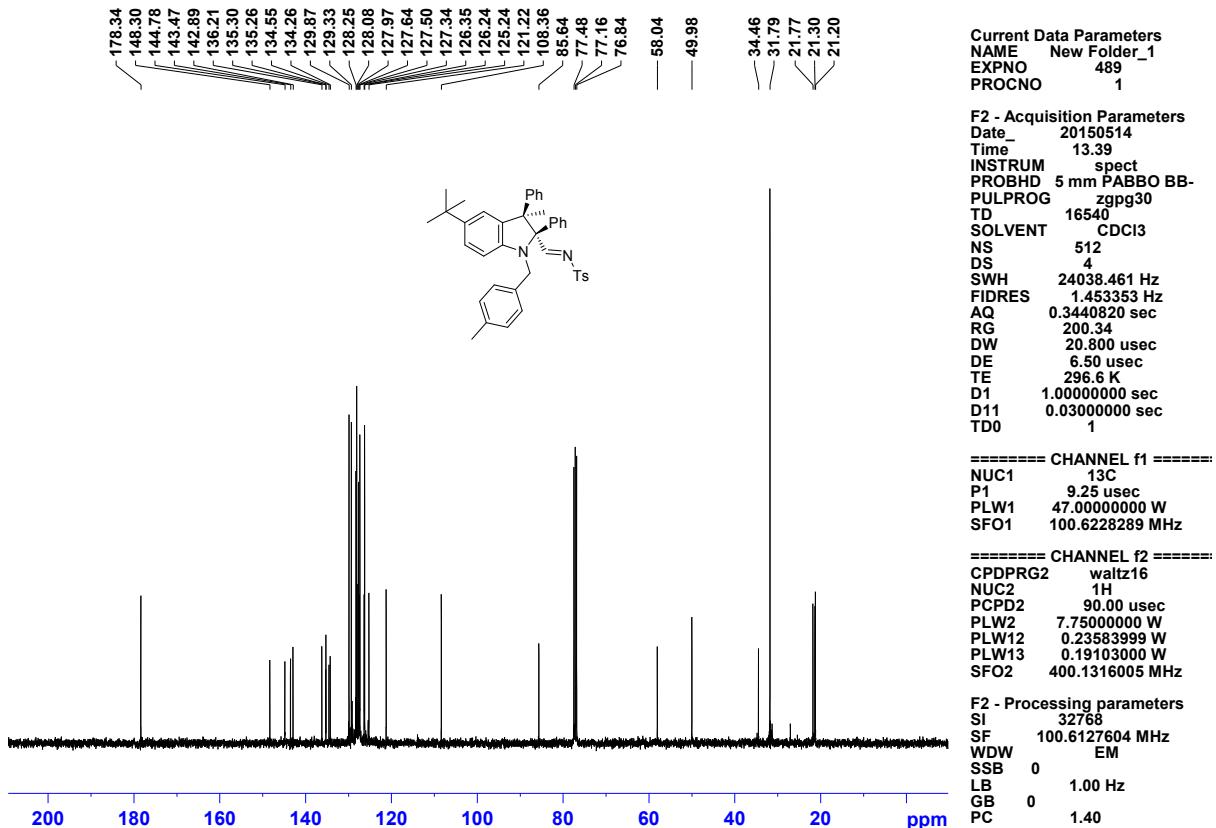


**Indoline (3ag):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

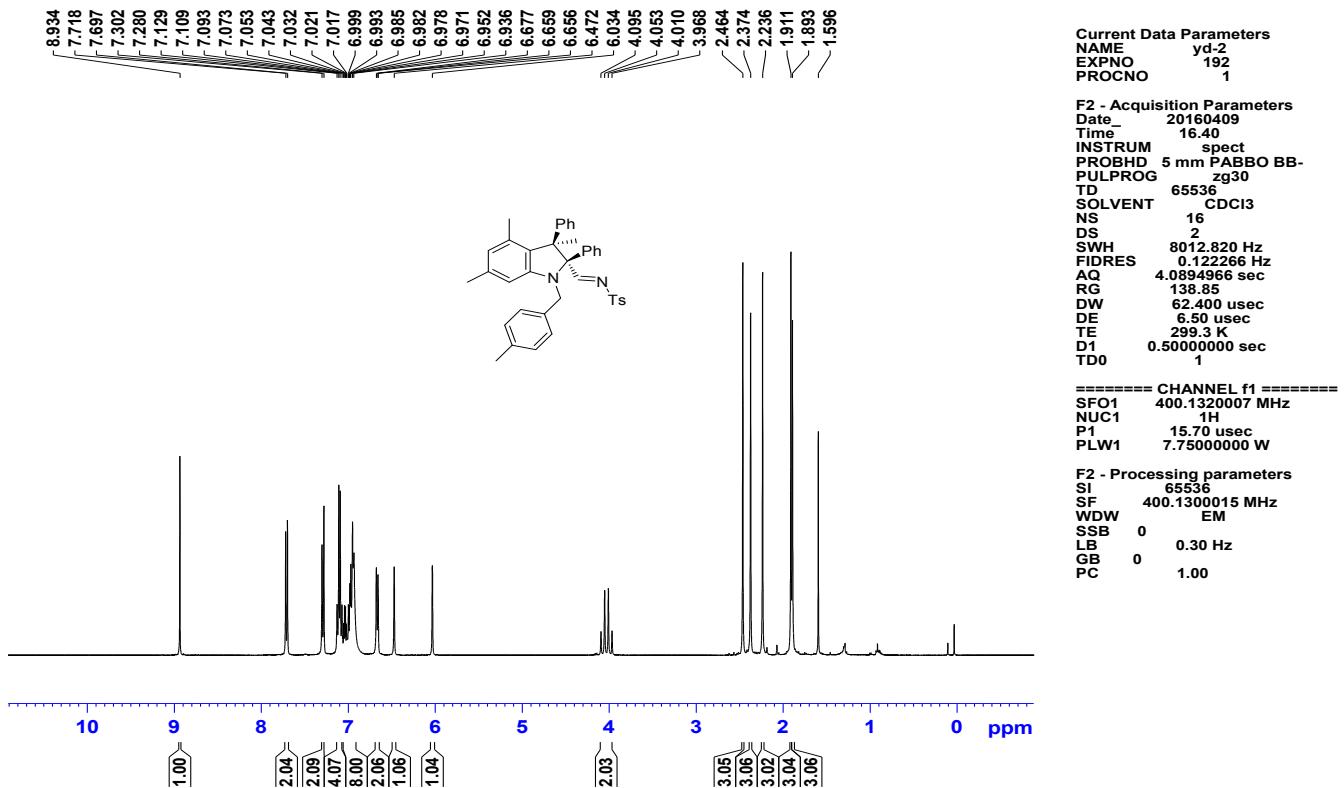


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

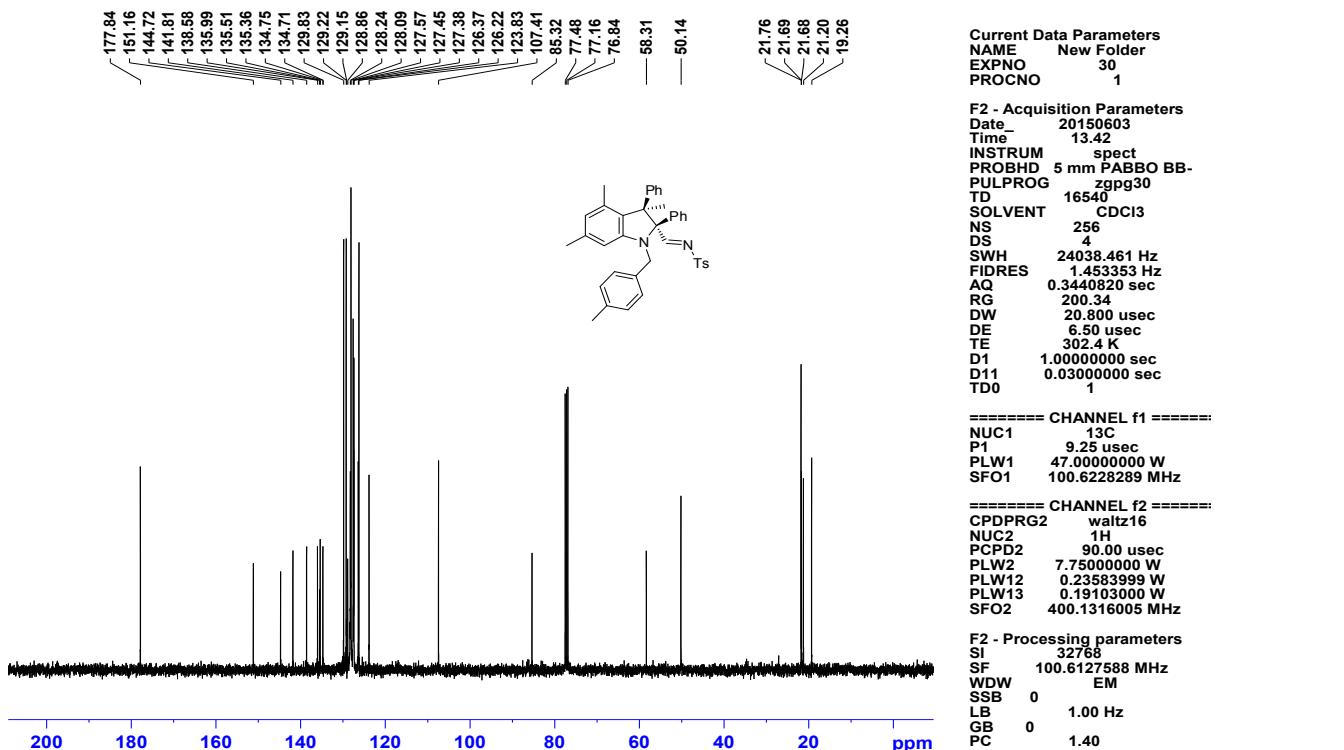


**Indoline (3ah):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

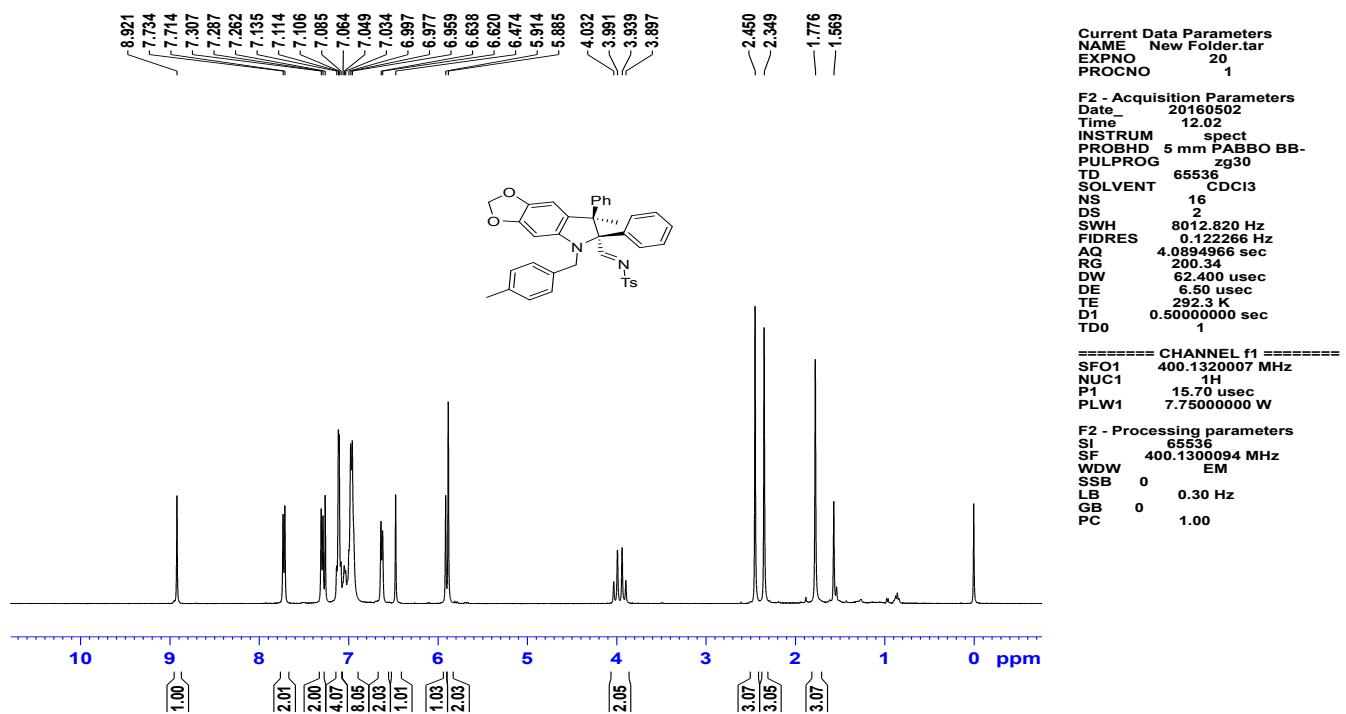


**$^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

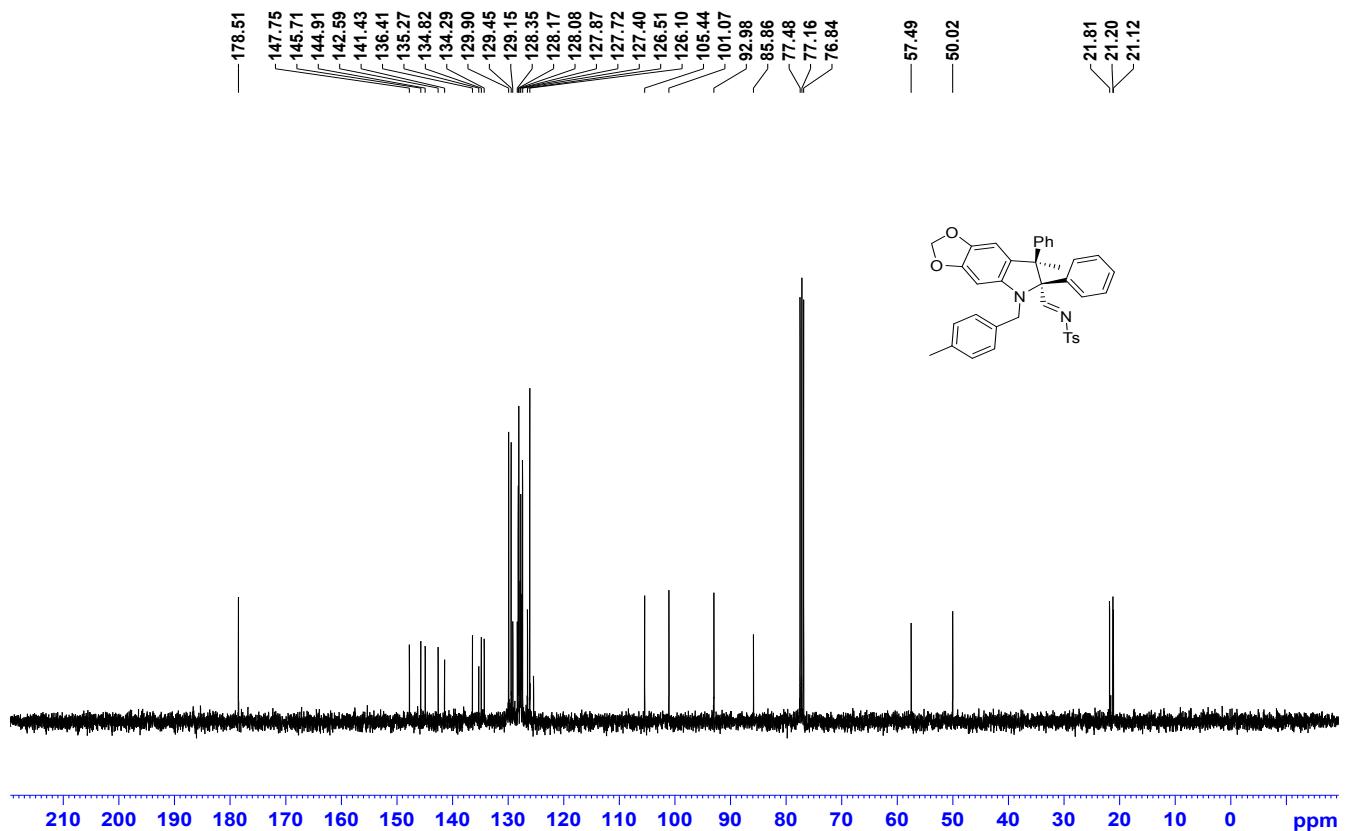


**Indoline (3ai):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

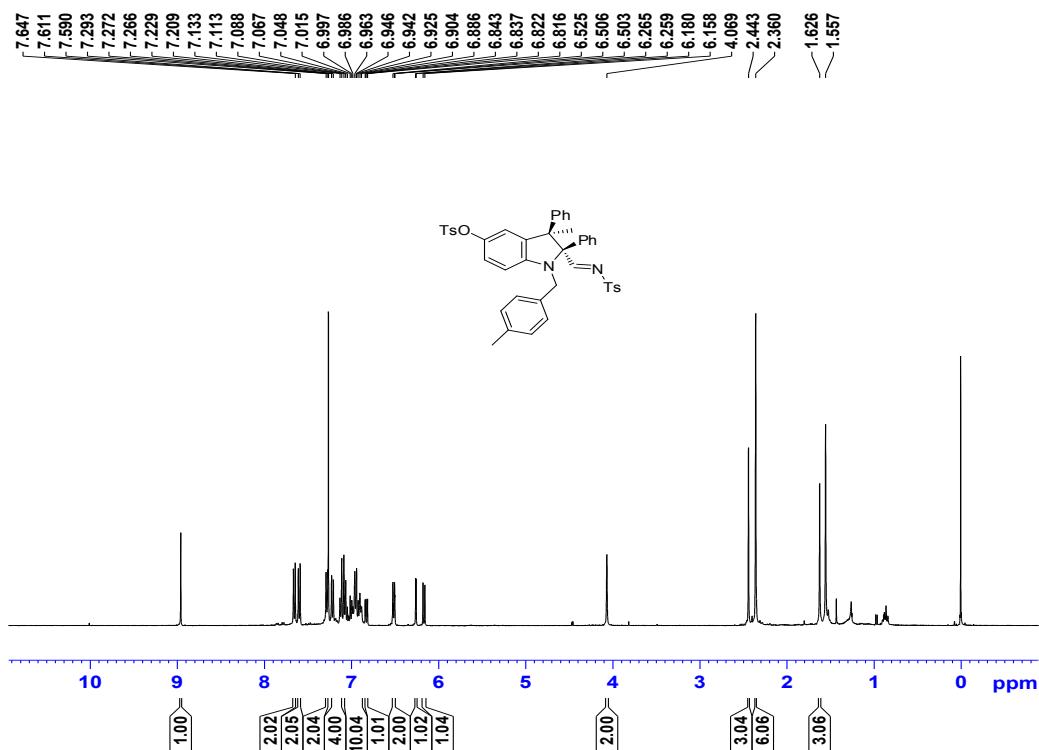


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

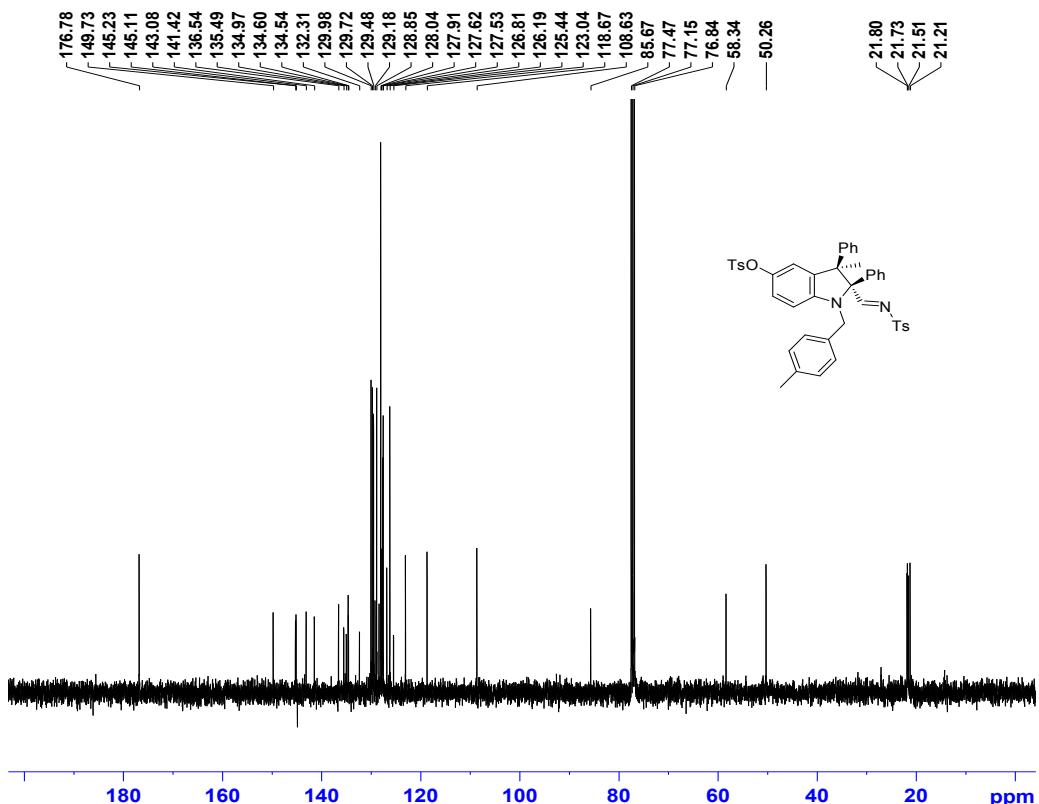


**Indoline (3aj):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

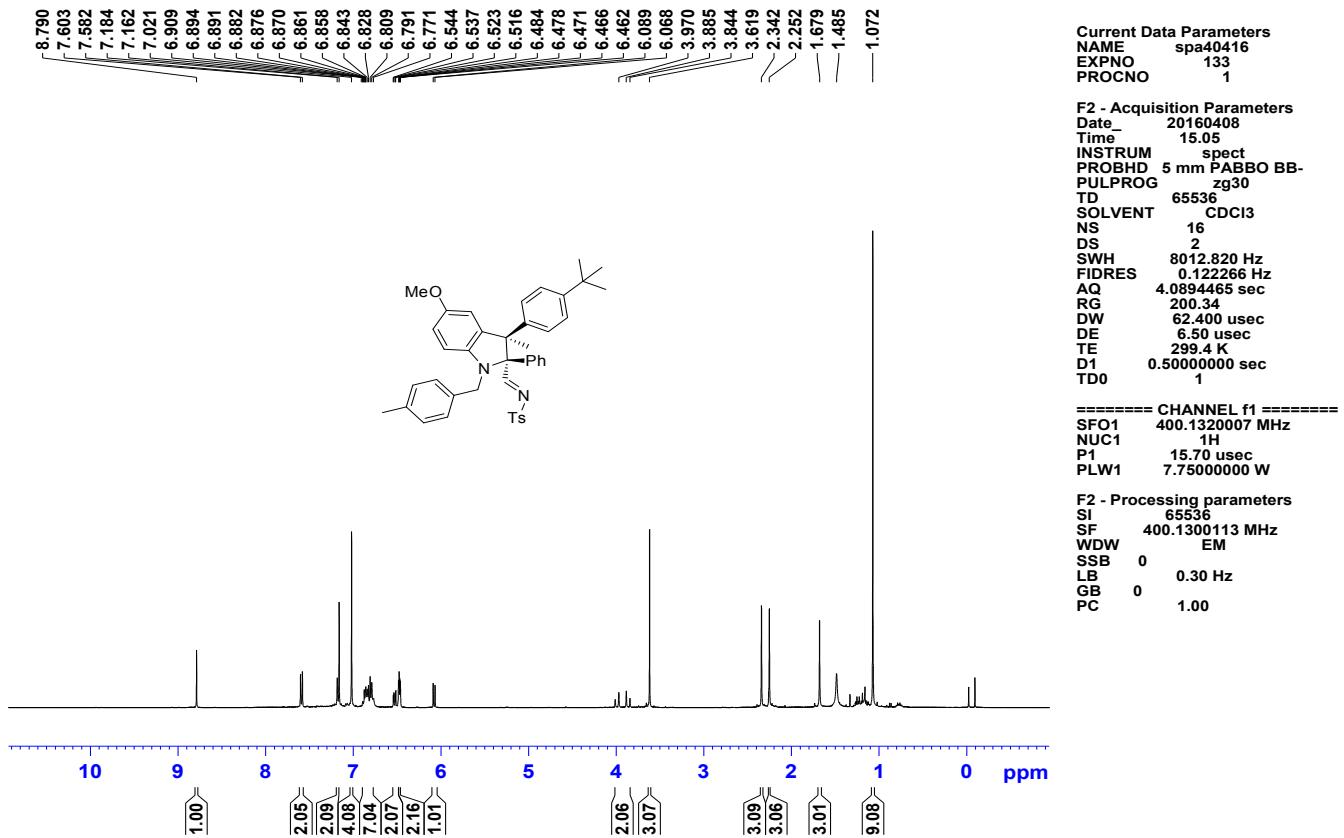


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

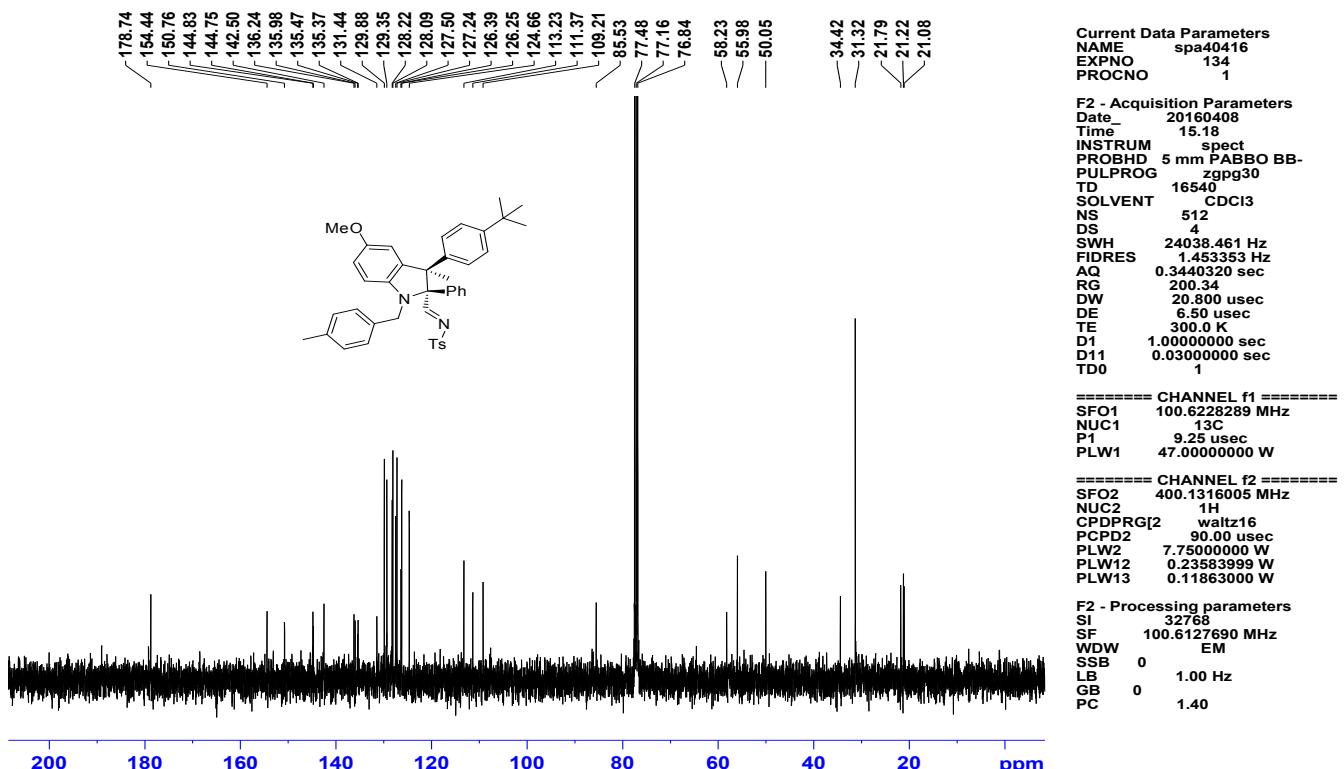


**Indoline (3al):**

<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C)

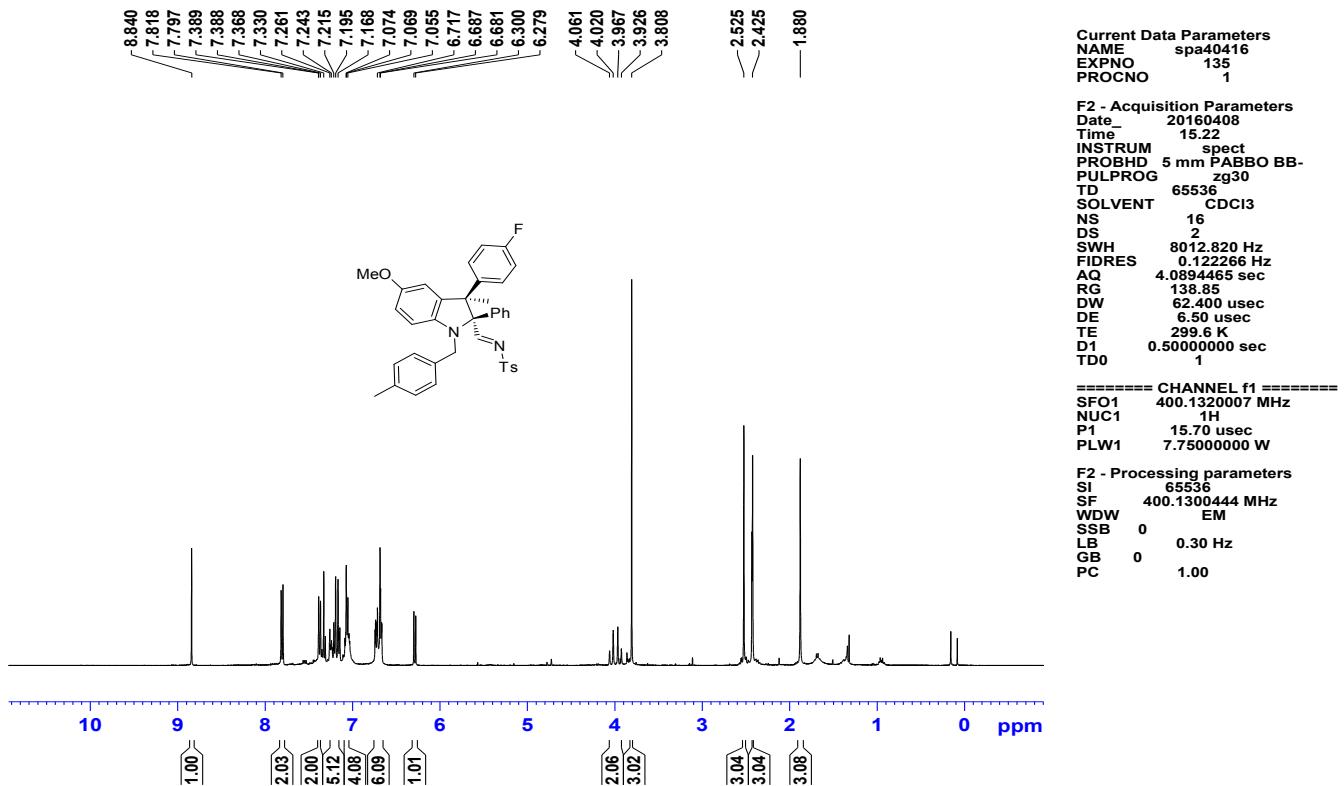


<sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C)

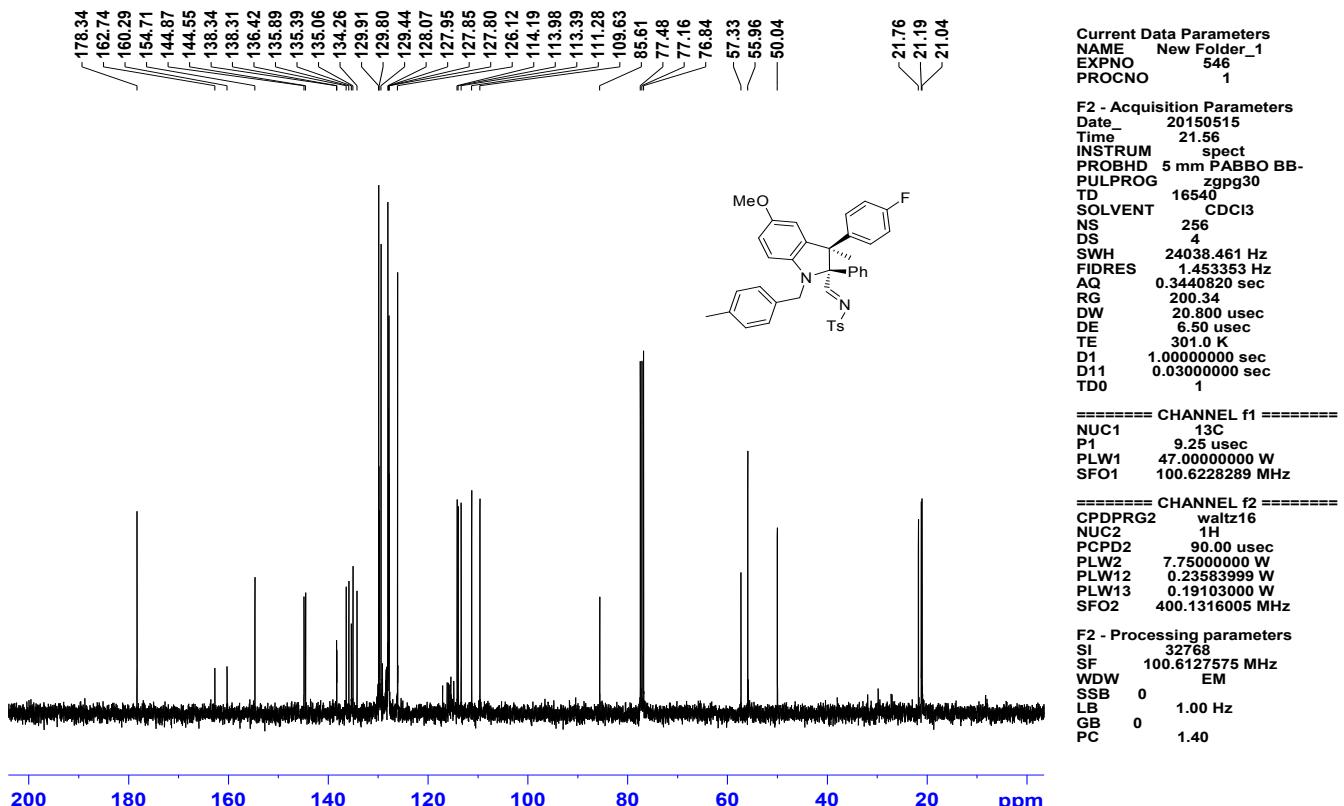


**Indoline (3am):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

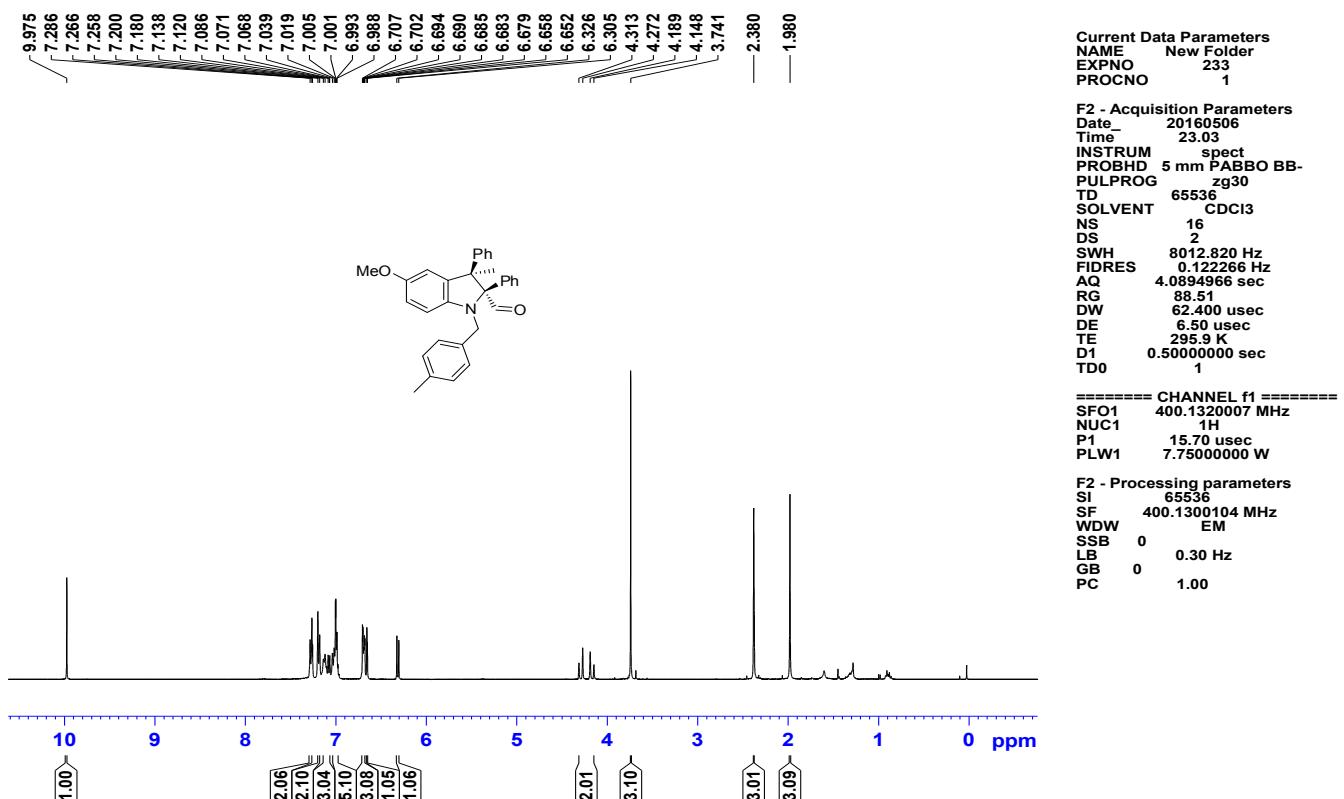


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

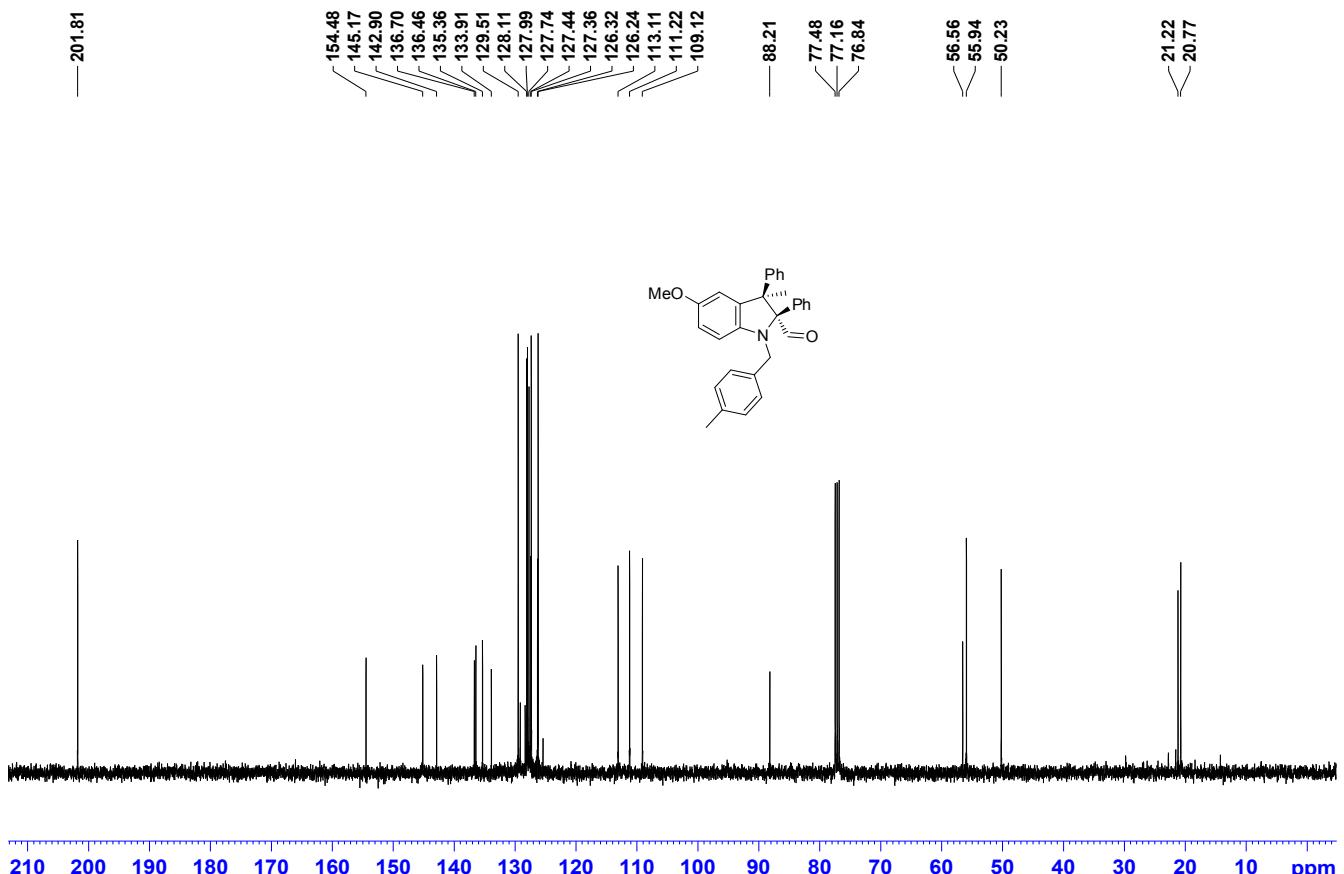


**Compound (4a):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

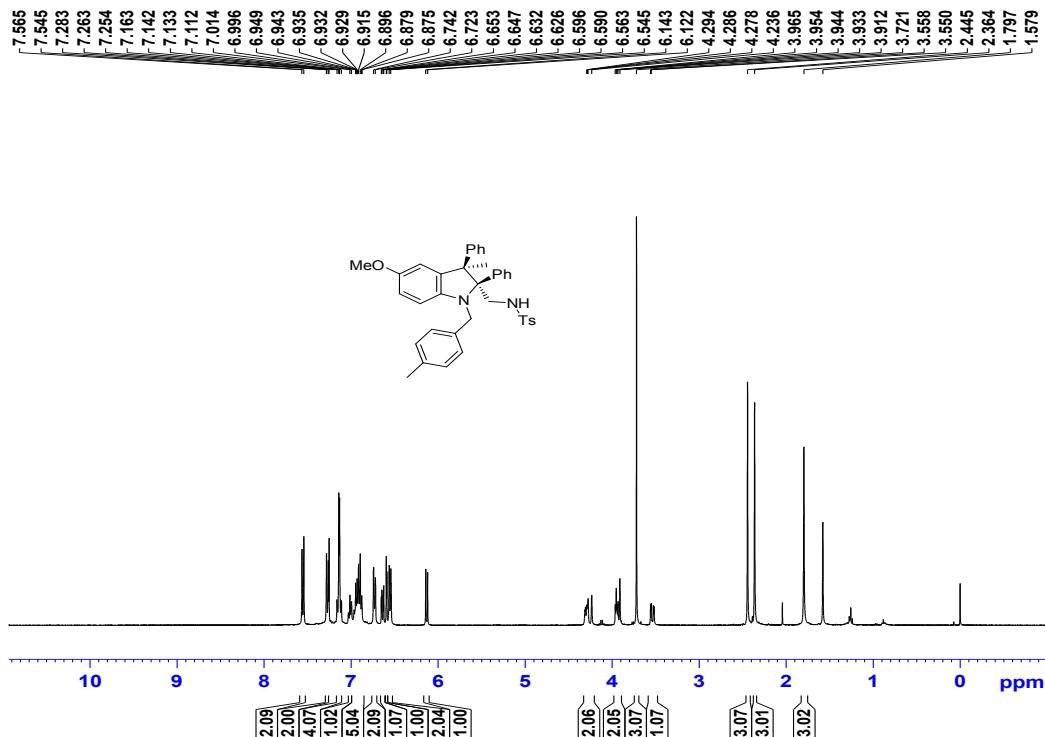


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

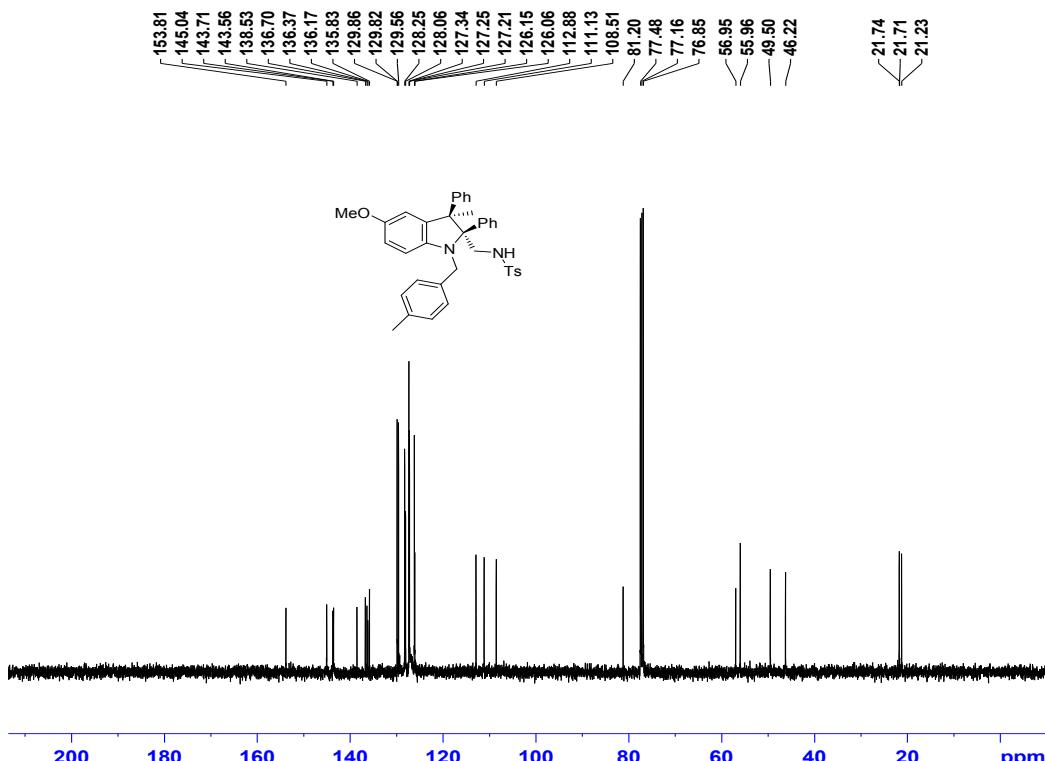


**Indoline (5a):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

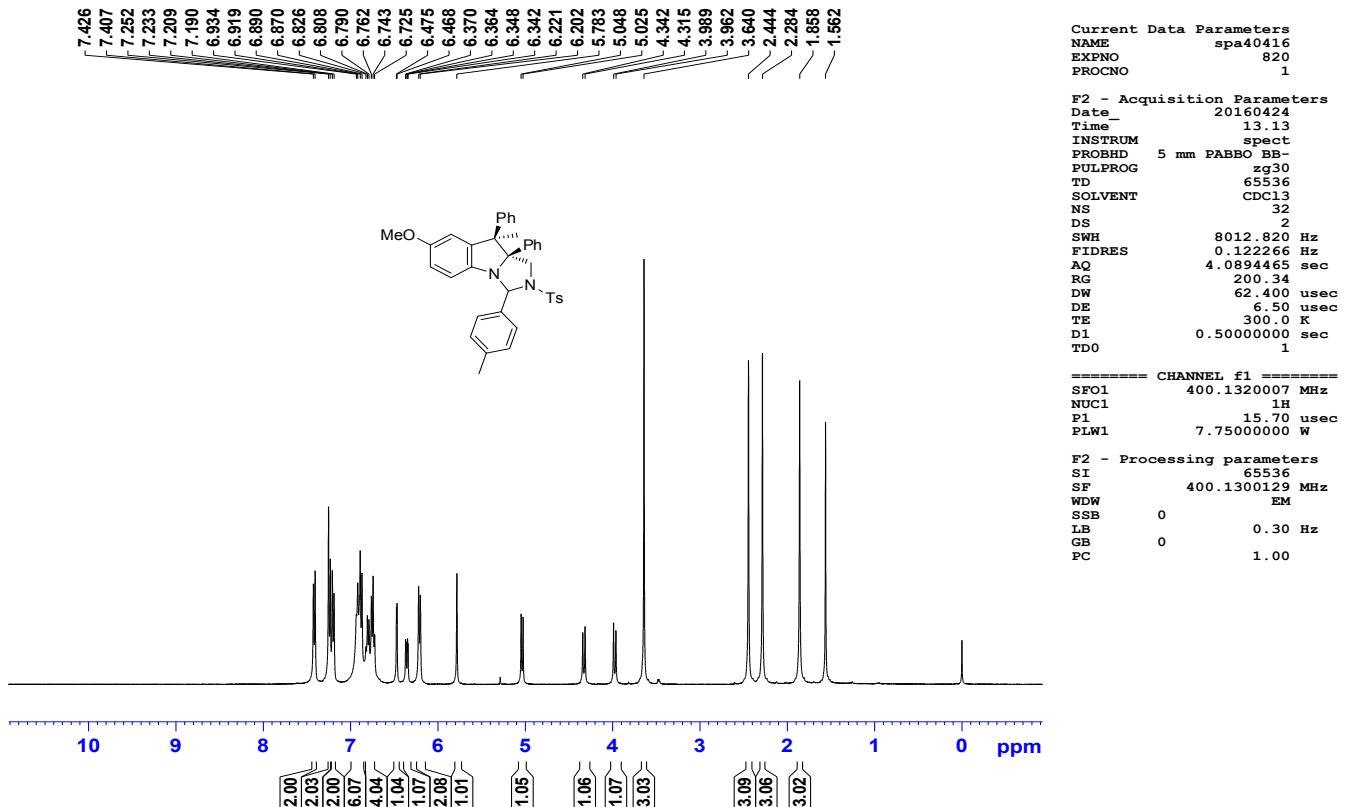


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

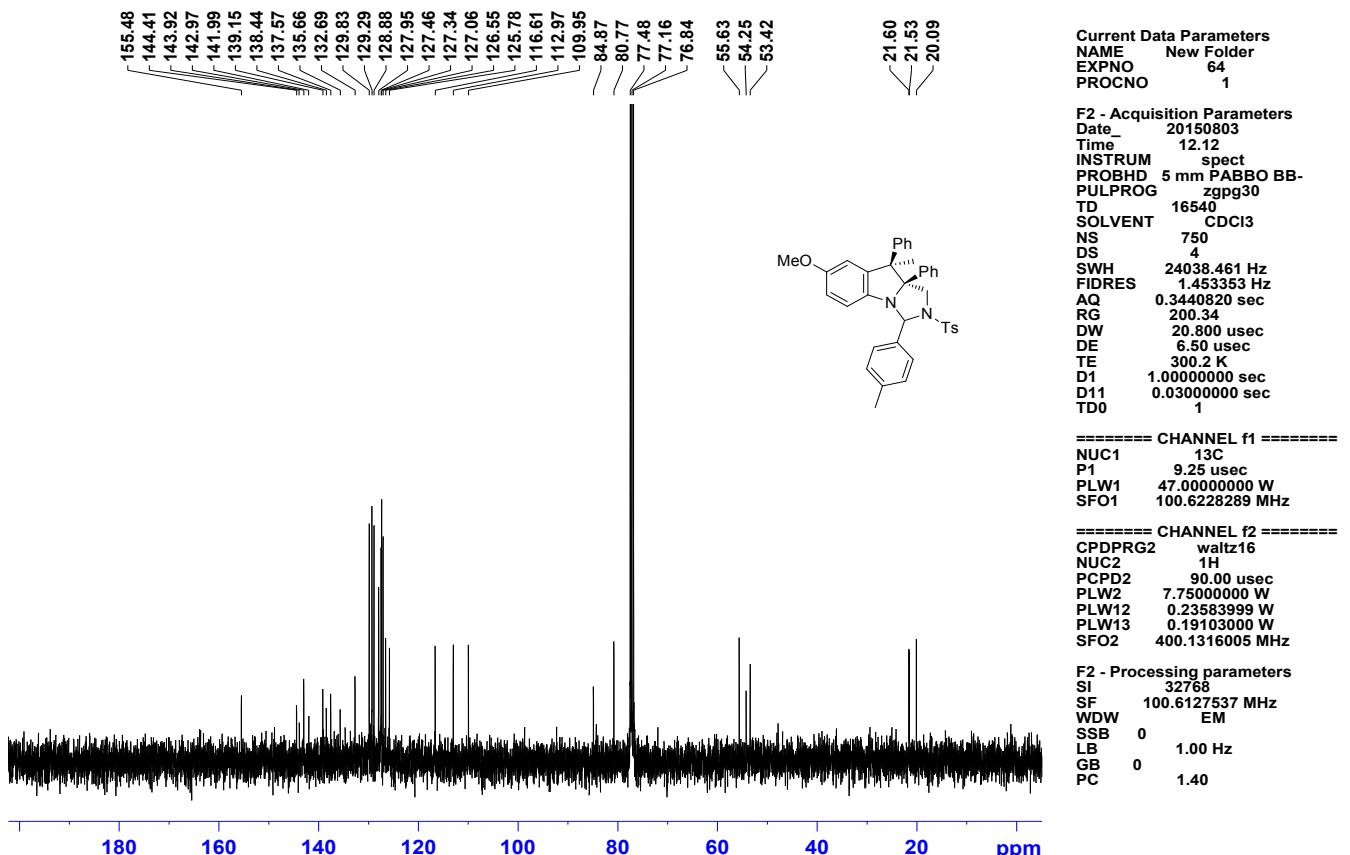


**Imidazoindoline (6a):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

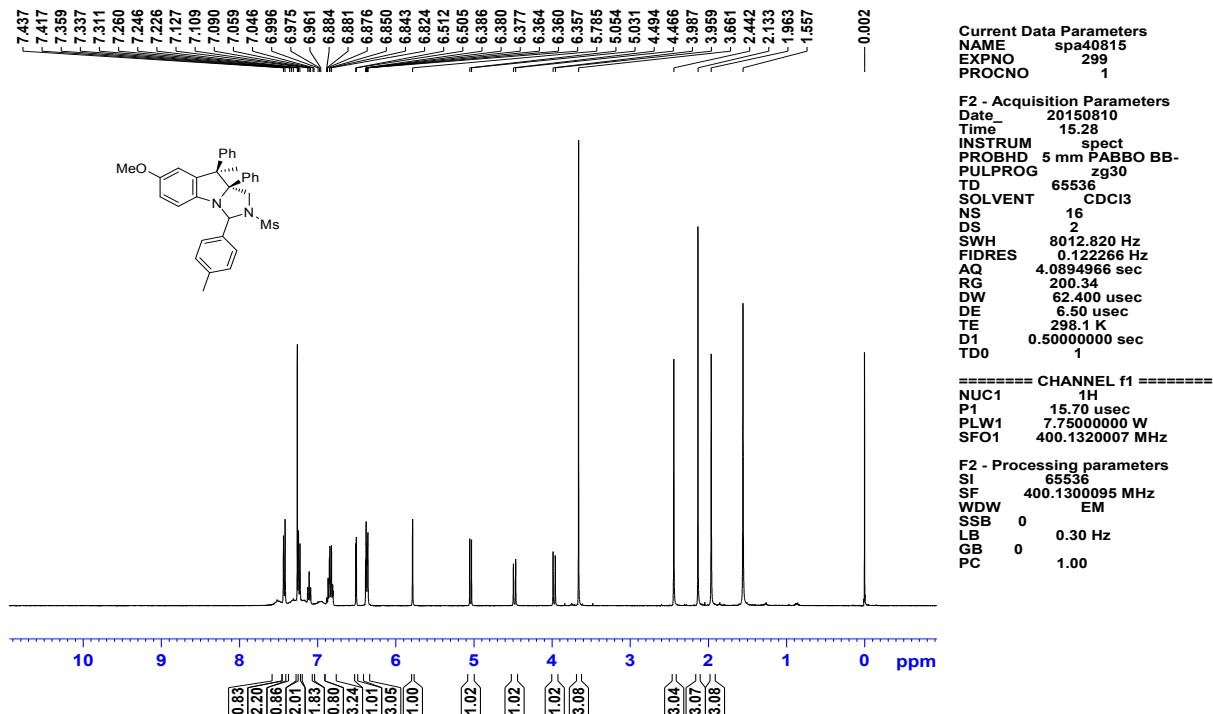


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

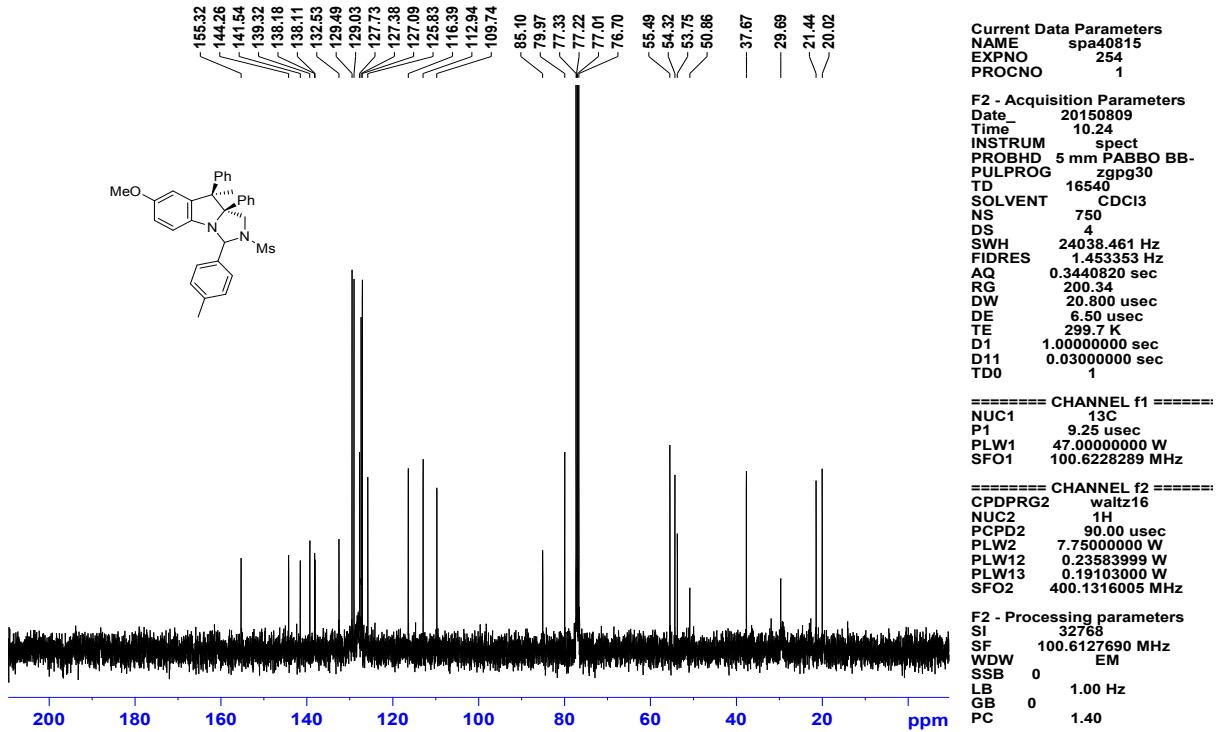


**Imidazoindoline (6b):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

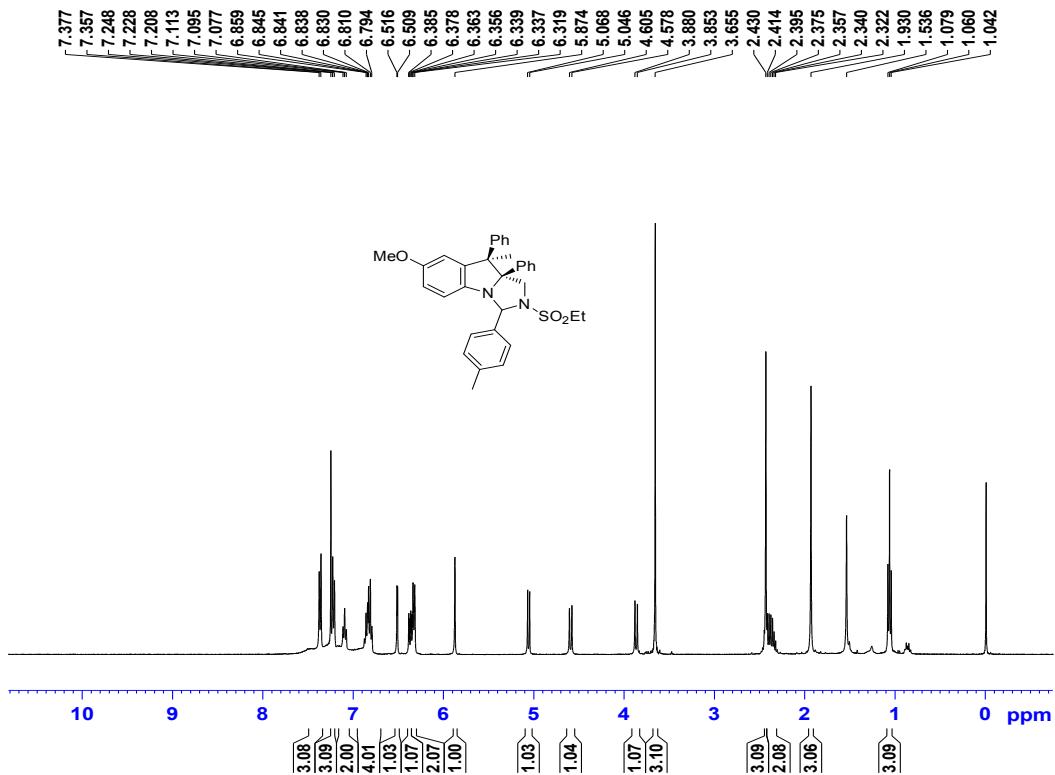


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



**Imidazoindoline (6c):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**



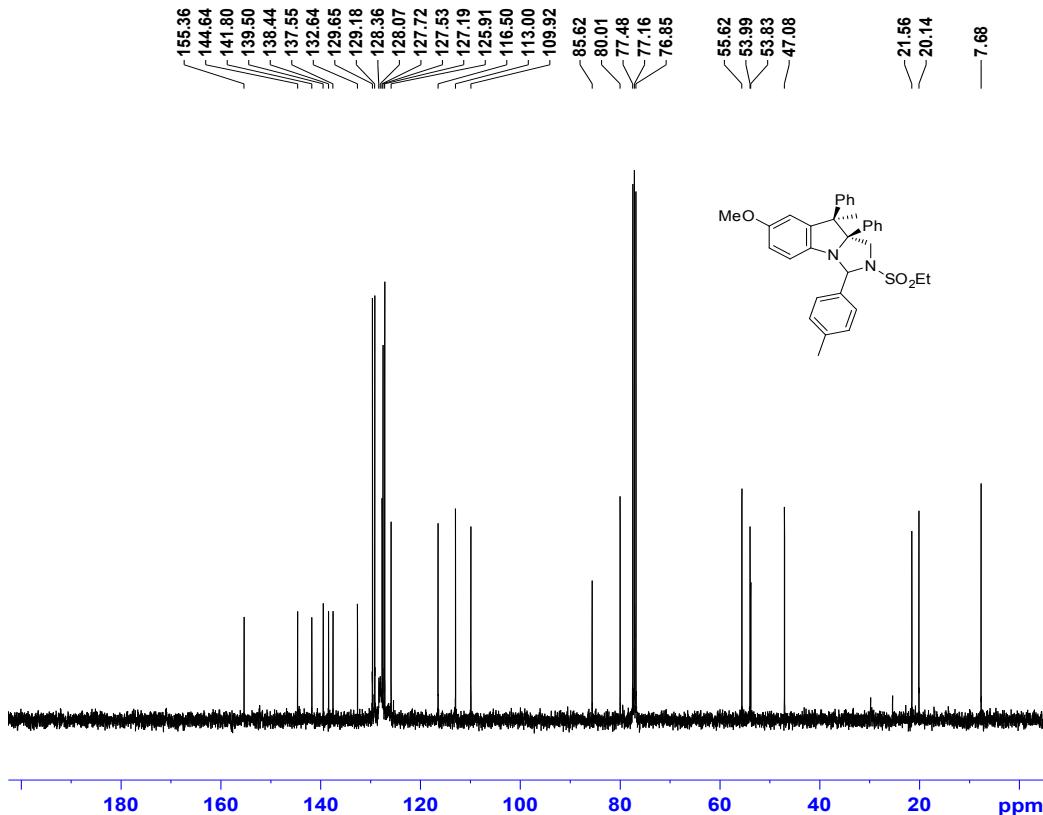
Current Data Parameters  
NAME yd-2  
EXPNO 712  
PROCNO 1

F2 - Acquisition Parameters  
Date\_ 20160422  
Time\_ 10.05  
INSTRUM spect  
PROBHD 5 mm PABBO BB-  
PULPROG zg30  
TD 65536  
SOLVENT CDCl<sub>3</sub>  
NS 16  
DS 2  
SWH 8012.820 Hz  
FIDRES 0.122266 Hz  
AQ 4.0894966 sec  
RG 200.34  
DW 62.400 usec  
DE 6.500 usec  
TE 299.9 K  
D1 0.5000000 sec  
TD0 1

===== CHANNEL f1 ======  
SFO1 400.1320007 MHz  
NUC1 1H  
P1 15.70 usec  
PLW1 7.75000000 W

F2 - Processing parameters  
SI 65536  
SF 400.1300143 MHz  
WDW EM  
SSB 0  
LB 0.30 Hz  
GB 0  
PC 1.00

**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



Current Data Parameters  
NAME spa40815  
EXPNO 130  
PROCNO 1

F2 - Acquisition Parameters  
Date\_ 20150805  
Time\_ 18.21  
INSTRUM spect  
PROBHD 5 mm PABBO BB-  
PULPROG zgpg30  
TD 16540  
SOLVENT CDCl<sub>3</sub>  
NS 512  
DS 4  
SWH 24038.461 Hz  
FIDRES 1.4340820 sec  
AQ 0.3440820 sec  
RG 200.34  
DW 20.800 usec  
DE 6.500 usec  
TE 299.5 K  
D1 1.0000000 sec  
D11 0.03000000 sec  
TD0 1

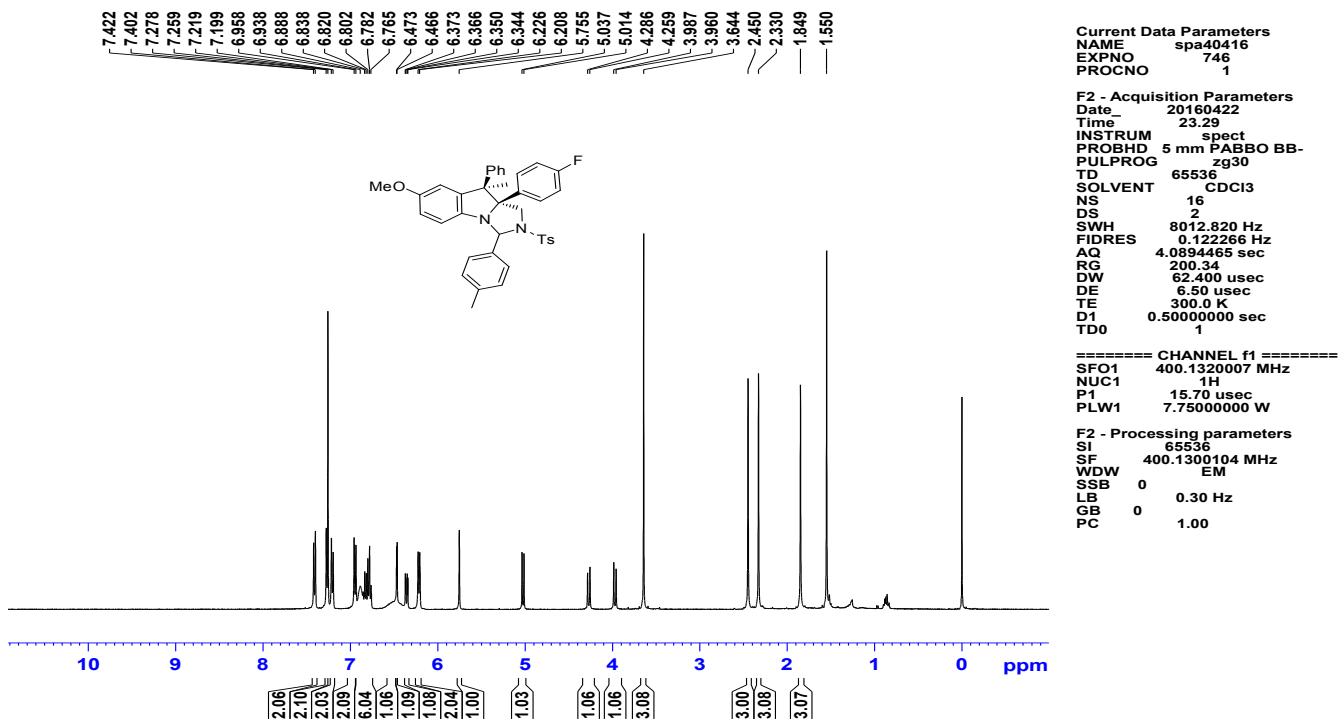
===== CHANNEL f1 ======  
NUC1 13C  
P1 9.25 usec  
PLW1 47.00000000 W  
SFO1 100.6228289 MHz

===== CHANNEL f2 ======  
CPDPRG2 waltz16  
NUC1 1H  
PCPD2 90.00 usec  
PLW2 7.75000000 W  
PLW12 0.23583999 W  
PLW13 0.19103000 W  
SFO2 400.1316005 MHz

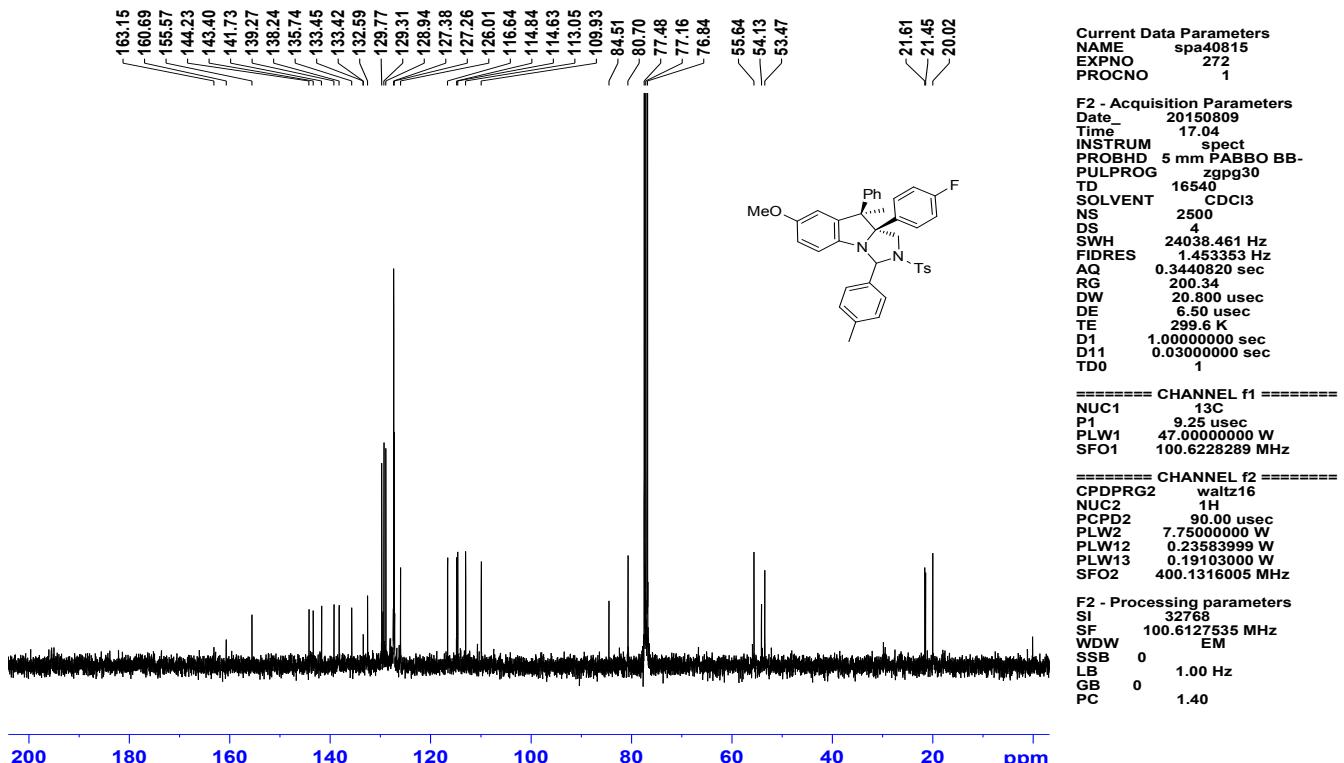
F2 - Processing parameters  
SI 32768  
SF 100.6127564 MHz  
WDW EM  
SSB 0  
LB 1.00 Hz  
GB 0  
PC 1.40

**Imidazoindoline (6d):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

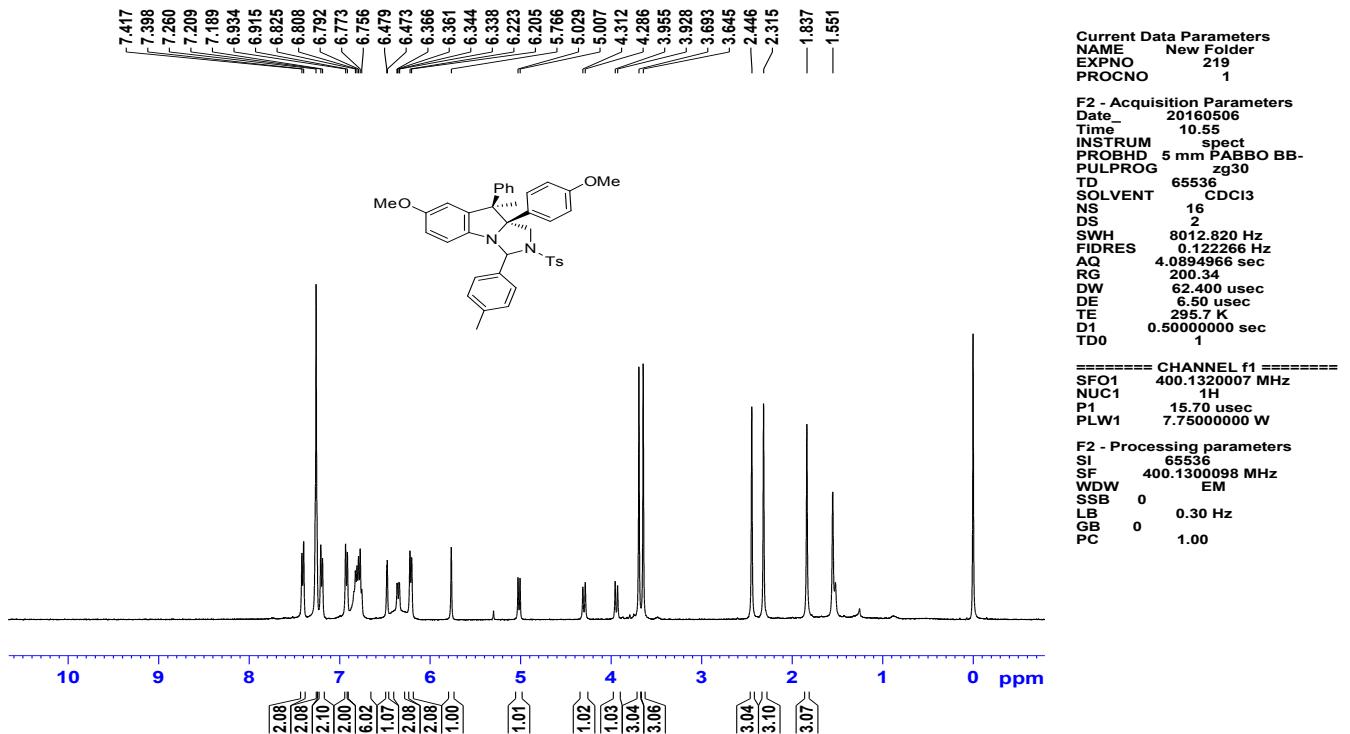


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

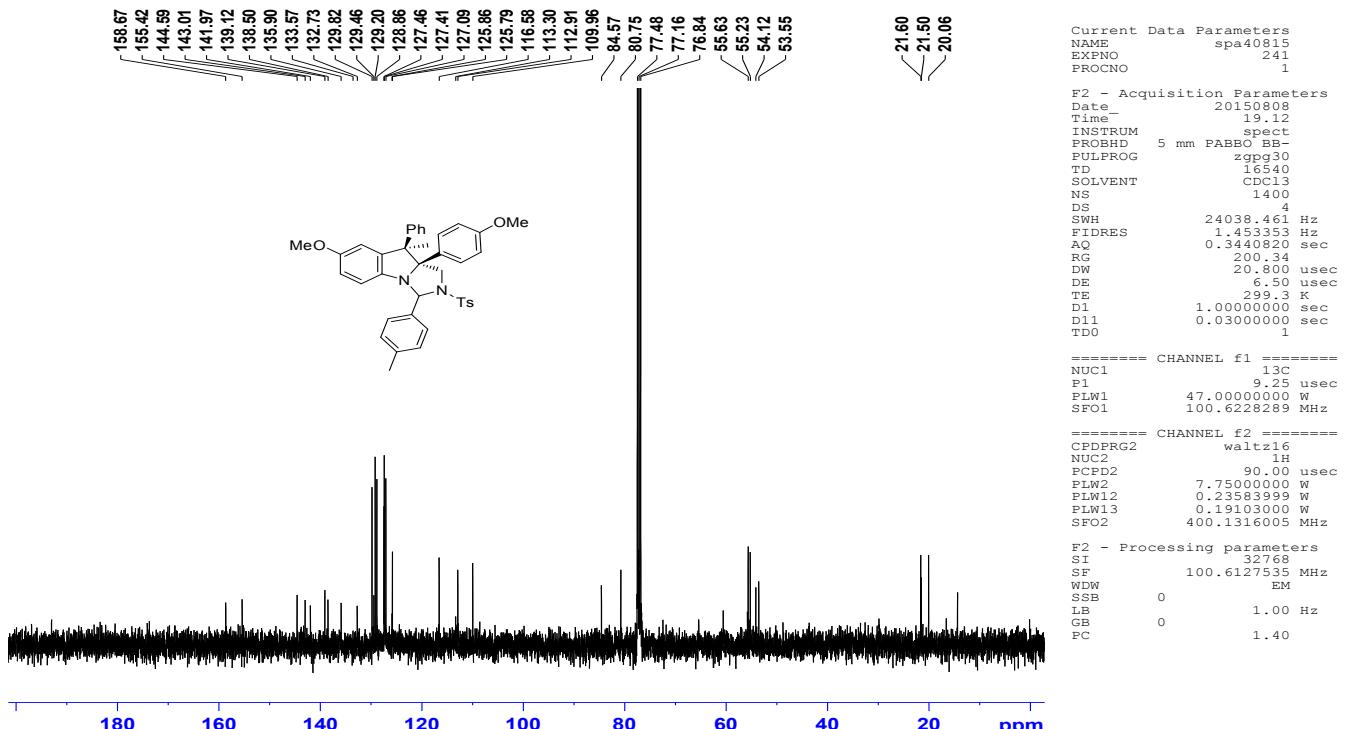


**Imidazoindoline (6e):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

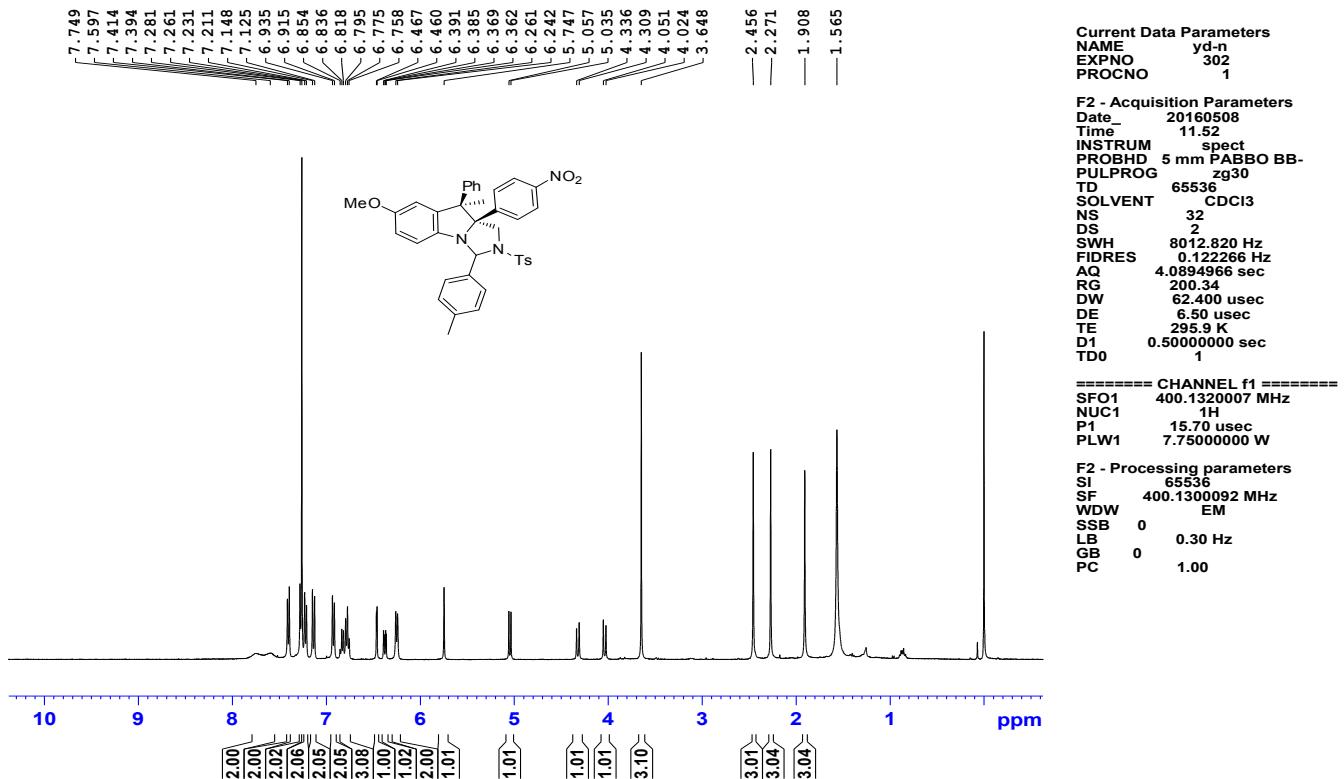


**$^{13}\text{C}\{\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

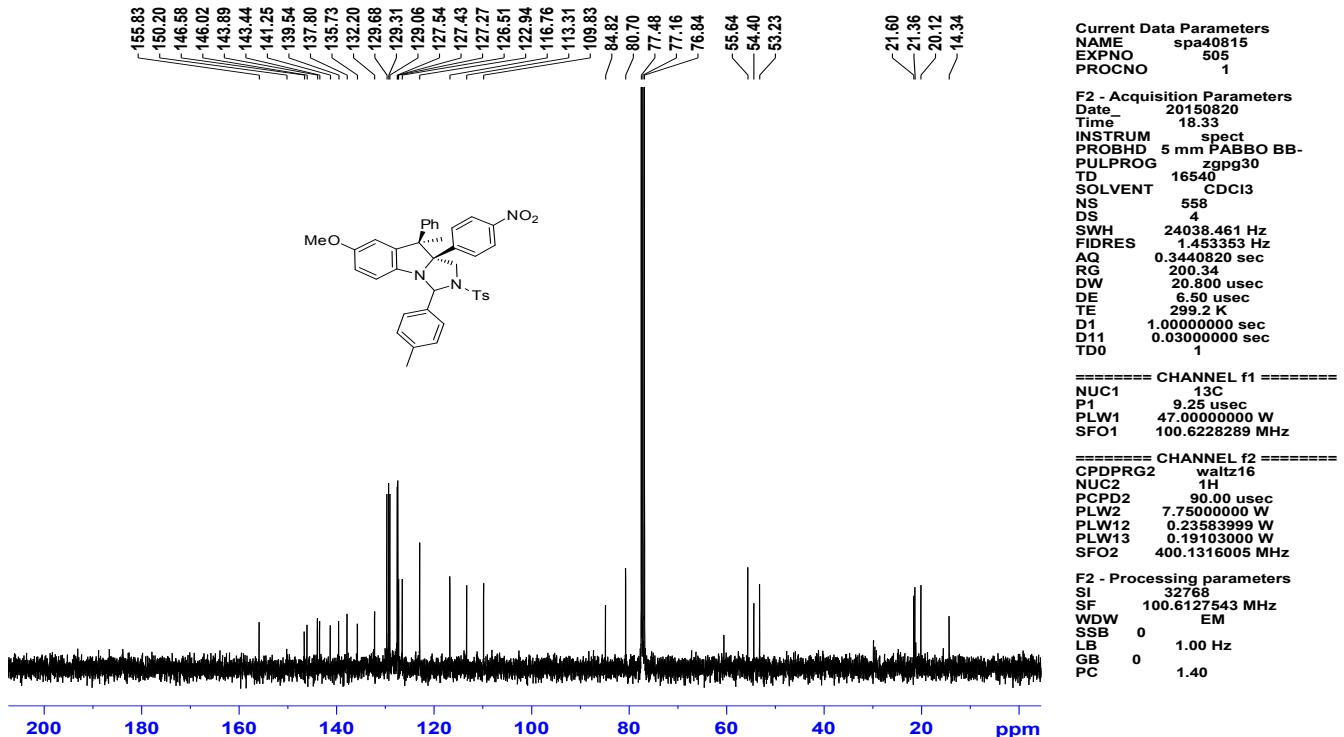


**Imidazoindoline (6f):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

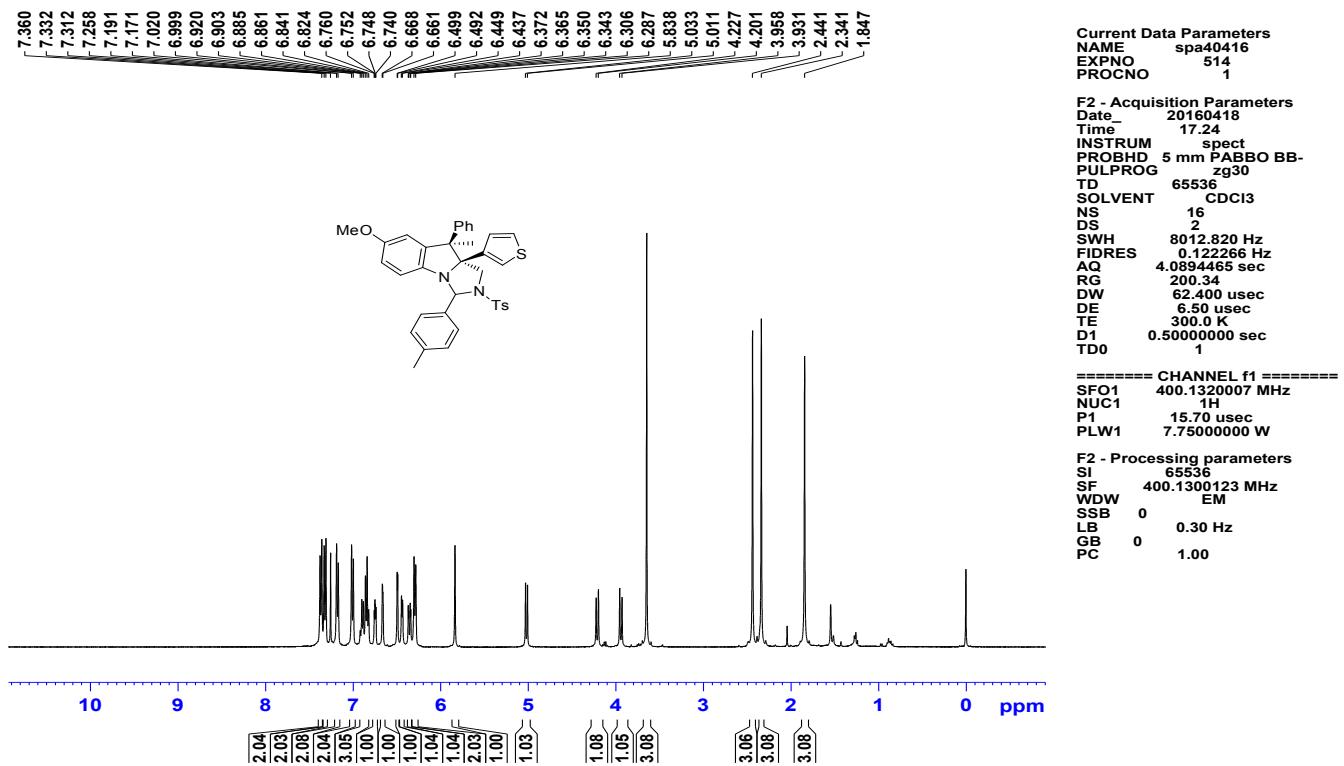


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

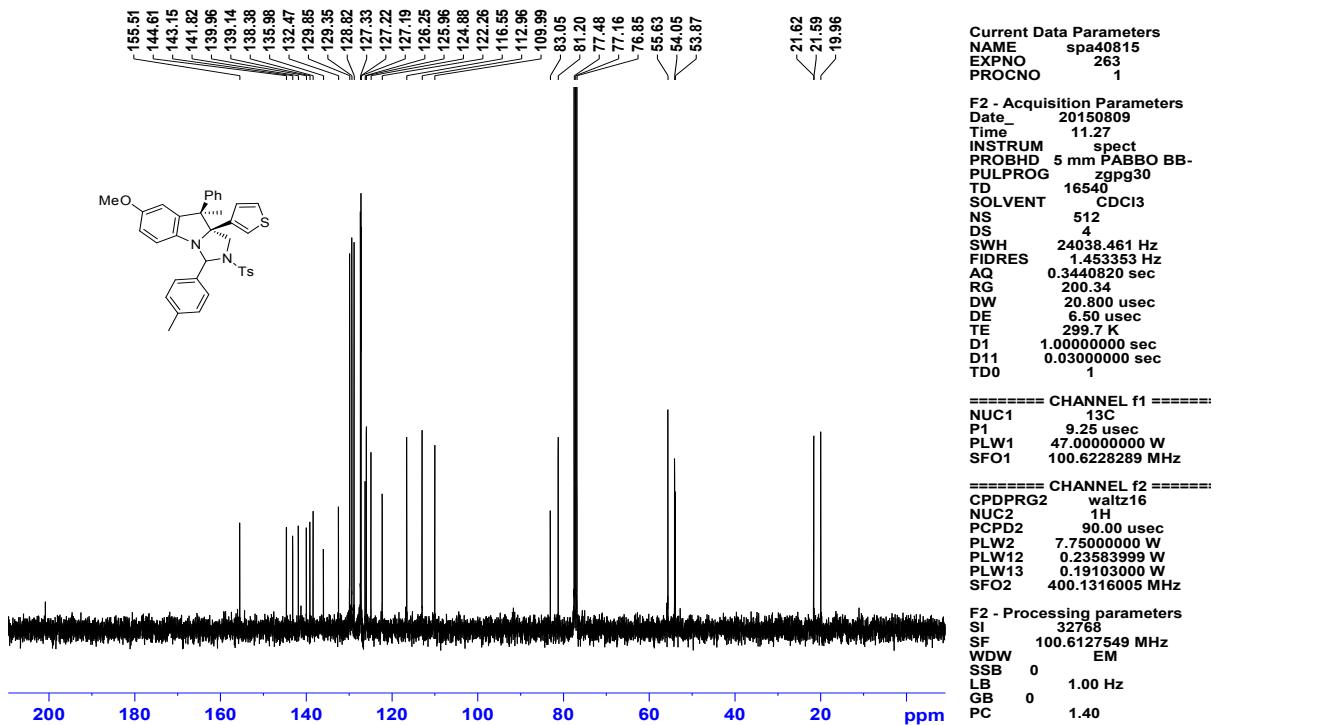


**Imidazoindoline (6g):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

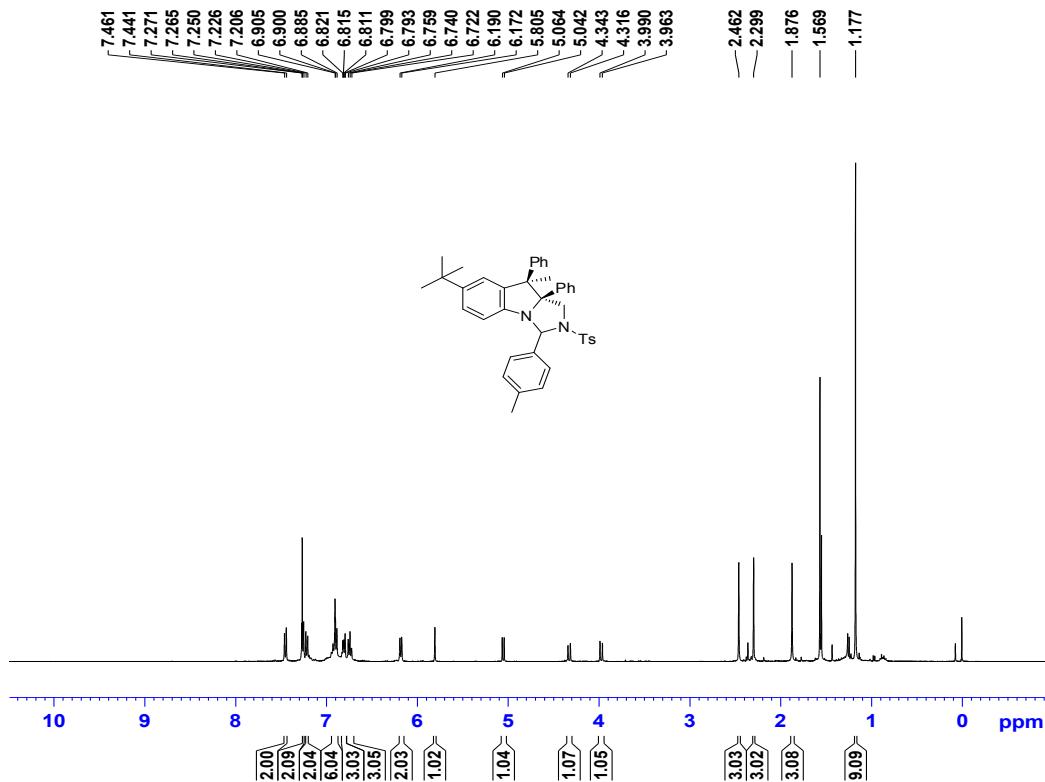


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



**Imidazoindoline (6h):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**



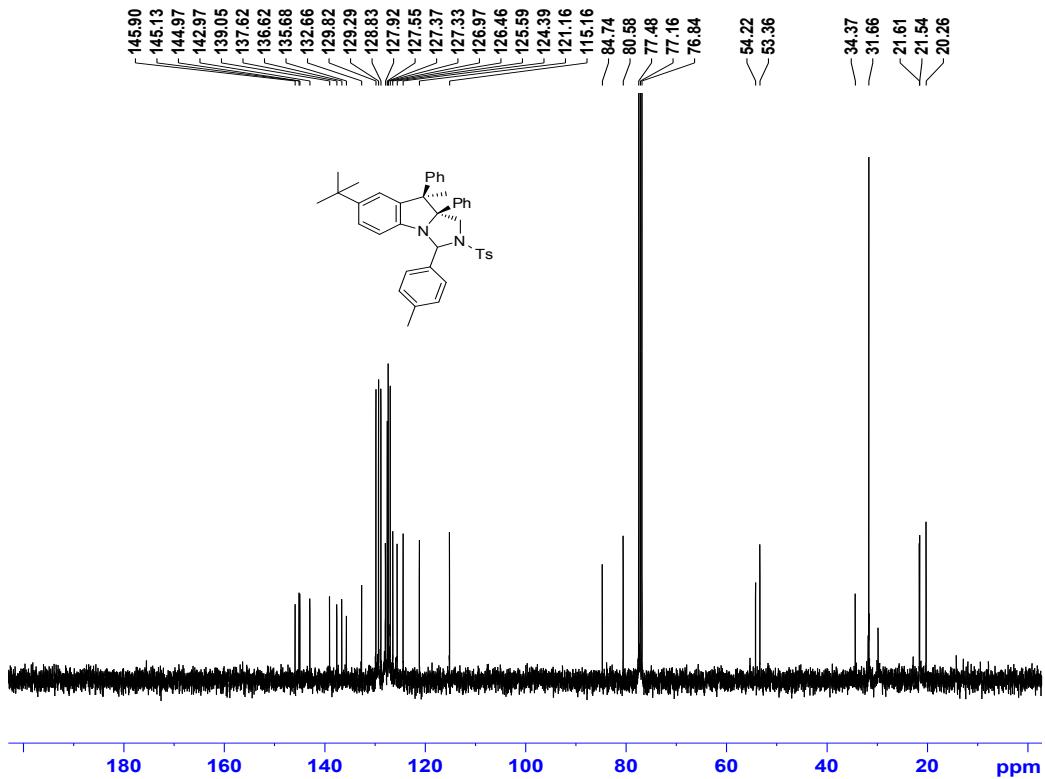
Current Data Parameters  
NAME spa40416  
EXPNO 132  
PROCNO 1

F2 - Acquisition Parameters  
Date 20160408  
Time 14.58  
INSTRUM spect  
PROBHD 5 mm PABBO BB-  
PULPROG zg30  
TD 65536  
SOLVENT CDCl<sub>3</sub>  
NS 16  
DS 2  
SWH 8012.820 Hz  
FIDRES 0.122266 Hz  
AQ 4.0894465 sec  
RG 200.34  
DW 62.400 usec  
DE 6.500 usec  
TE 299.4 K  
D1 0.5000000 sec  
TDO 1

===== CHANNEL f1 ======  
SFO1 400.1320007 MHz  
NUC1 1H  
P1 15.70 usec  
PLW1 7.75000000 W

F2 - Processing parameters  
SI 65536  
SF 400.1300108 MHz  
WDW EM  
SSB 0  
LB 0.30 Hz  
GB 0  
PC 1.00

**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**



Current Data Parameters  
NAME spa40815  
EXPNO 229  
PROCNO 1

F2 - Acquisition Parameters  
Date 20150808  
Time 15.47  
INSTRUM spect  
PROBHD 5 mm PABBO BB-  
PULPROG zgpg30  
TD 16540  
SOLVENT CDCl<sub>3</sub>  
NS 512  
DS 4  
SWH 24038.461 Hz  
FIDRES 1.453353 Hz  
AQ 0.3440820 sec  
RG 200.34  
DW 20.800 usec  
DE 6.50 usec  
TE 299.1 K  
D1 1.0000000 sec  
D11 0.03000000 sec  
TDO 1

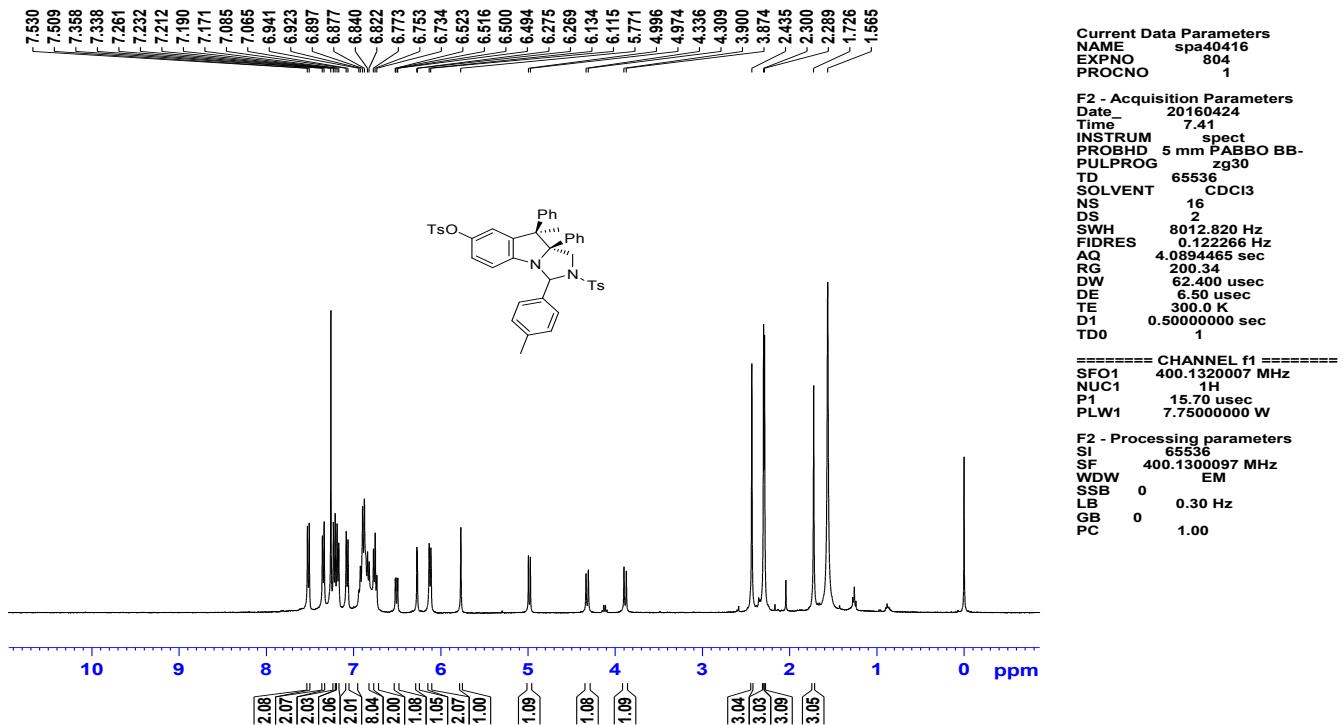
===== CHANNEL f1 ======  
NUC1 13C  
P1 9.25 usec  
PLW1 47.00000000 W  
SFO1 100.6228289 MHz

===== CHANNEL f2 ======  
CPDPRG2 waltz16  
NUC2 1H  
PCPD2 90.00 usec  
PLW2 7.75000000 W  
PLW12 0.23583999 W  
PLW13 0.19103000 W  
SFO2 400.1316005 MHz

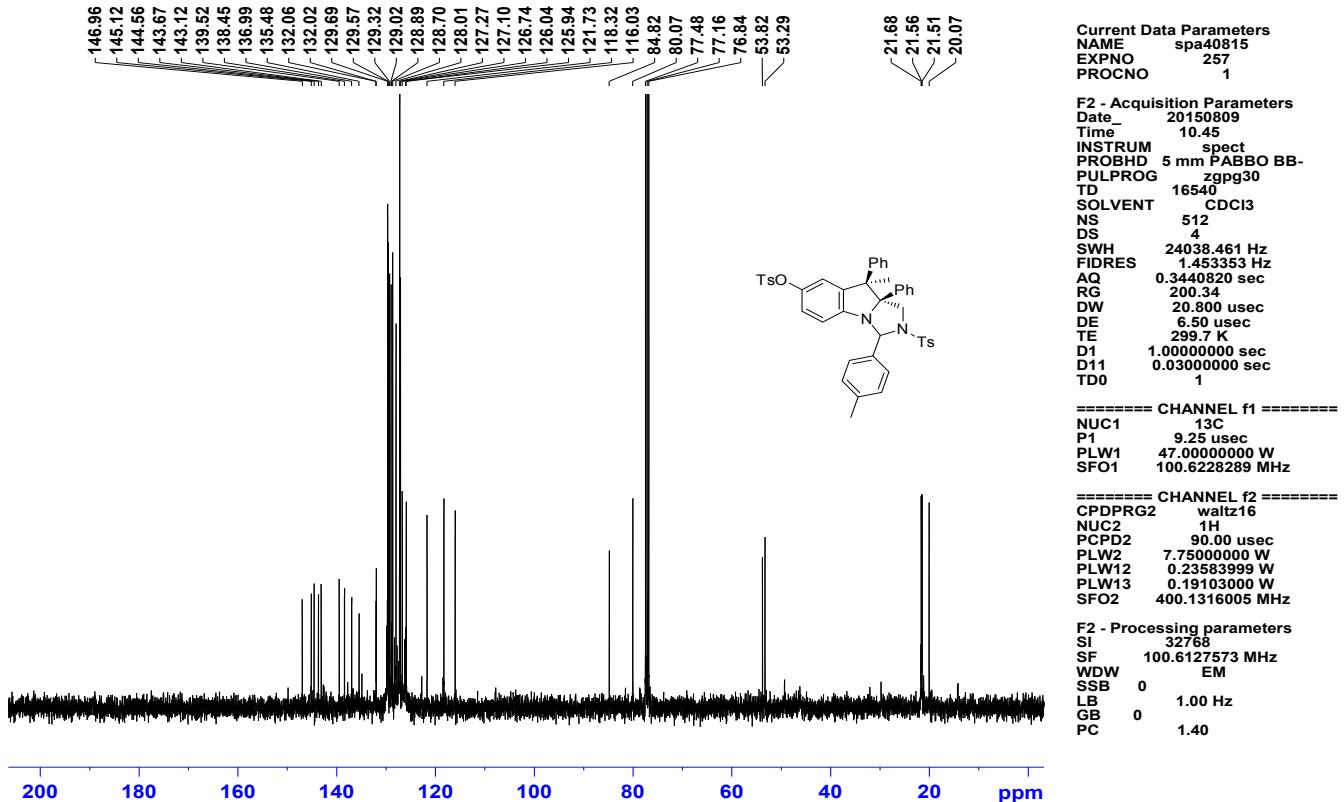
F2 - Processing parameters  
SI 32768  
SF 100.6127547 MHz  
WDW EM  
SSB 0  
LB 1.00 Hz  
GB 0  
PC 1.40

**Imidazoindoline (6i):**

**$^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ , 24 °C)**

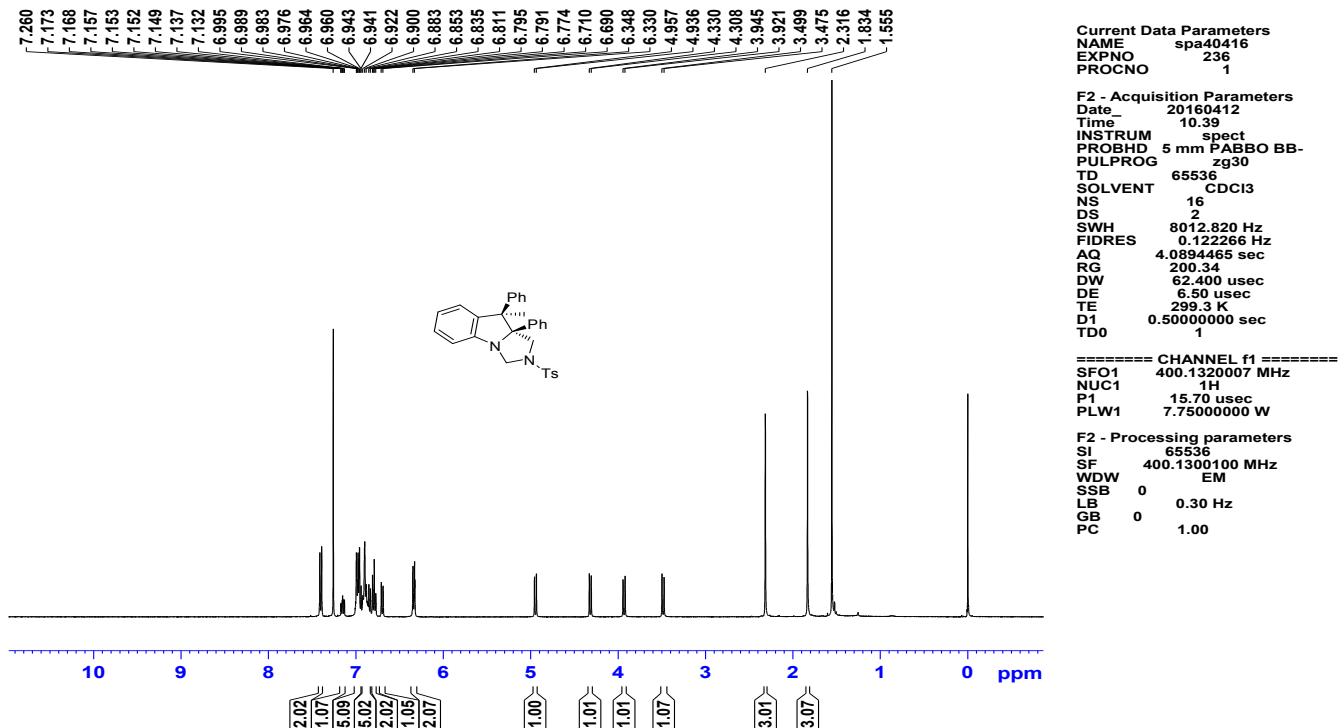


**$^{13}\text{C}\{^1\text{H}\}$  NMR (100 MHz,  $\text{CDCl}_3$ , 24 °C)**

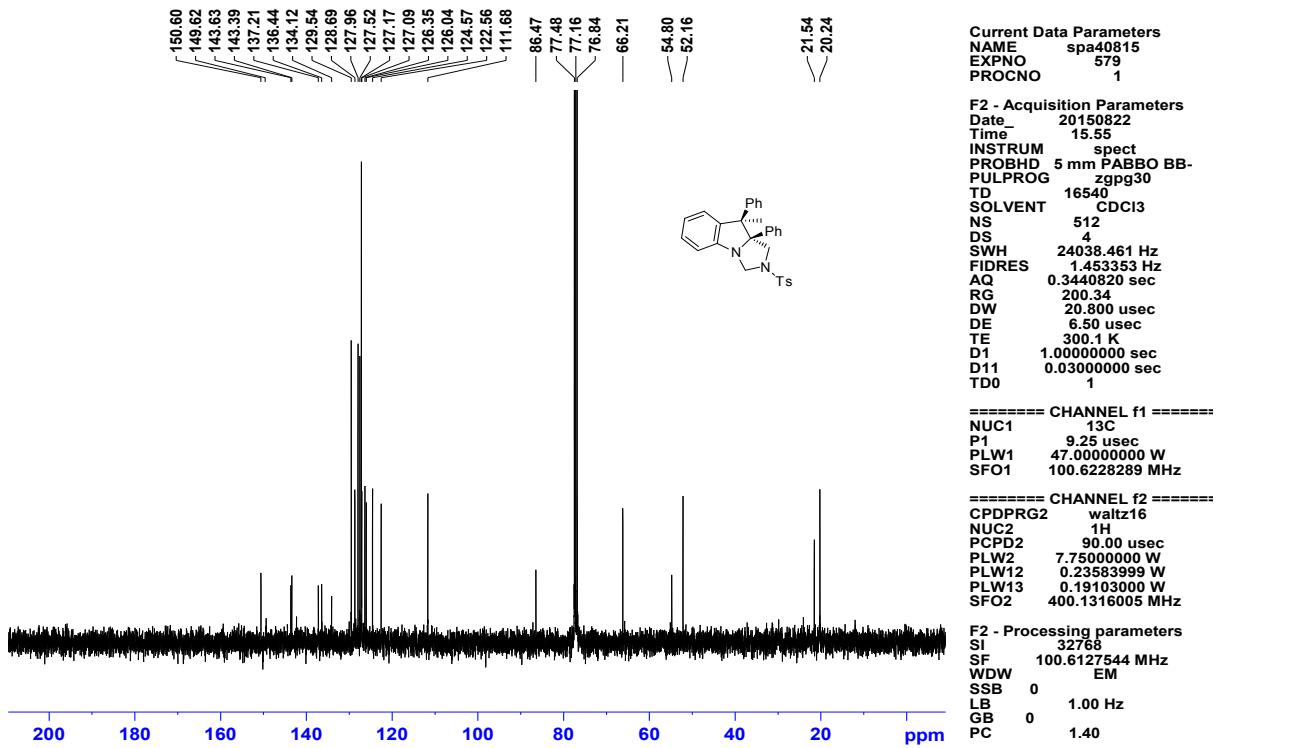


**Imidazoindoline (6k):**

**<sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>, 24 °C)**



**<sup>13</sup>C{<sup>1</sup>H} NMR (100 MHz, CDCl<sub>3</sub>, 24 °C)**



## 12. NOE experiment:

