

## Supporting Information

### **Ruthenium(II)-Catalyzed C–H Activation/C–N Bond Formation via in situ Generated Iminophosphorane as the Directing Group: Construction of Annulated Pyridin-2(1*H*)-ones**

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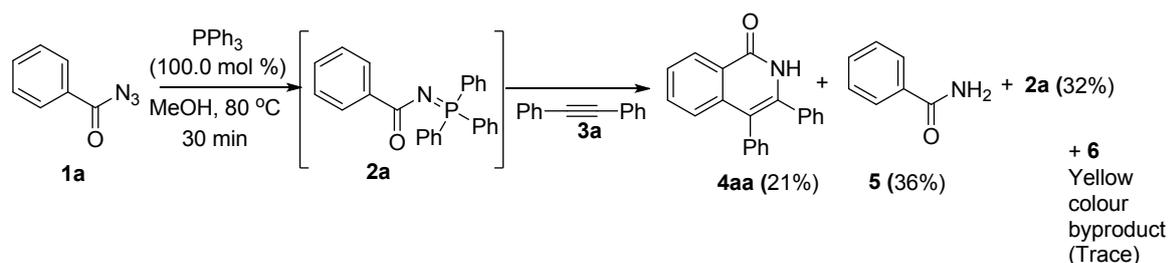
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## Experimental Section

### I. Mechanistic observation reactions

#### A Isolation of intermediates iminophosphorane and benzamide during the transformation of **1a** in MeOH

Scheme 1. Quenching the reaction after 5h

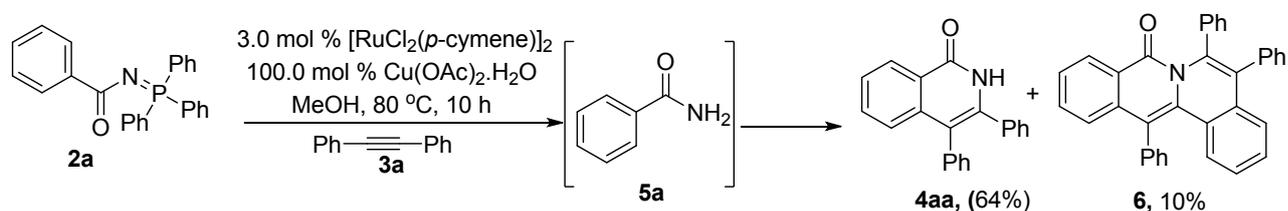


<sup>a</sup> Reaction condition: (1.02 mmol) of **1a**, (0.81 mmol) of **3a**, 3.0 mol %  $[\text{Ru}(p\text{-cymene})\text{Cl}_2]_2$ , (1.02 mmol) of  $\text{Cu}(\text{OAc})_2 \cdot \text{H}_2\text{O}$  in 5 mL of MeOH at 80 °C after 5h.

In a flame dried round bottomed flask equipped with a stir bar, we added a mixture of Acyl azide **1a** (1.02 mmol) (150 mg), in 5.0 mL solvent followed by the addition of Triphenylphosphine (TPP) (1.02 mmol) (267 mg), we then monitored the formation of iminophosphorane **2a** by tlc after 30 min of stirring at 80 °C. Next we added alkyne **3a** (0.816 mmol) (145 mg),  $[\text{RuCl}_2(p\text{-cymene})]_2$  (3.0 mol%) and  $\text{Cu}(\text{OAc})_2 \cdot \text{H}_2\text{O}$  (1.02 mmol) (204 mg). After 5 h of reaction monitored by tlc. The resulting mixture was evaporated to remove MeOH. After that diluted with  $\text{H}_2\text{O}$  (10 mL) and mixture was extracted with EtOAc (3x20 mL). The combined organic layers were washed with brine (20 mL) and dried over  $\text{Na}_2\text{SO}_4$ . The solvent was evaporated in vacuo and the remaining residue was purified by column chromatography on silica gel (*n*-hexanes/EtOAc 5%-60%) to yield products **4aa** (21%, 63 mg), benzamide **5a** (36%, 44 mg), unreacted iminiophosphorane **2a** (32%, 124 mg) along with traces of yellow colour byproduct **6**.

#### B. Treatment of presynthesized iminophosphorane **2a** with unactivated internal alkyne **3a** in MeOH

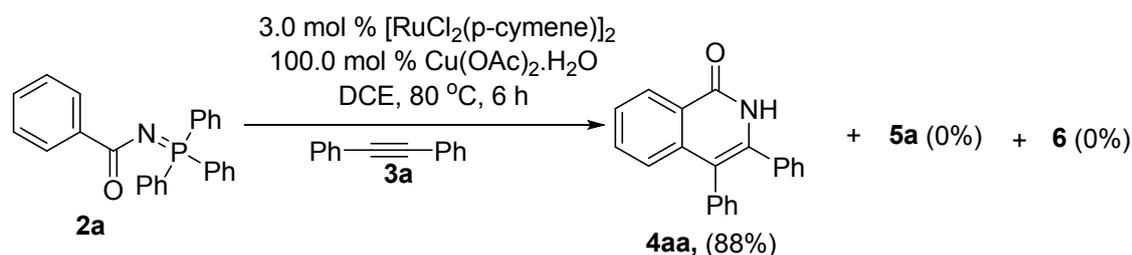
**Scheme 2:** Treatment of presynthesized iminophosphorane **2a** with unactivated internal alkyne **3a** in MeOH for 10 h



In a flame dried round bottomed flask equipped with a stir bar, we added a mixture of iminophosphorane **2a** (0.39 mmol) (150 mg), in 5.0 mL solvent followed by the addition of alkyne **3** (0.312 mmol),  $[\text{RuCl}_2(p\text{-cymene})]_2$  (3.0 mol%) and  $\text{Cu}(\text{OAc})_2 \cdot \text{H}_2\text{O}$  (0.39 mmol) (71 mg) at 80 °C. After completion of the reaction in 10 h, monitored by tlc, the resulting mixture was evaporated to remove MeOH. After that diluted with  $\text{H}_2\text{O}$  (10 mL) and mixture was extracted with EtOAc (3x20 mL). The combined organic layers were washed with brine (20 mL) and dried over  $\text{Na}_2\text{SO}_4$ . The solvent was evaporated in vacuo and the remaining residue was purified by column chromatography on silica gel (*n*-hexanes/EtOAc 5%-40%) to yield products **4aa** (64%, 74 mg) and **6** (10%, 18 mg).

### C. Treatment of presynthesized iminophosphorane **2a** with unactivated internal alkyne **3a** in DCE

**Scheme 3.** Synthesis of **4aa** from iminophosphorane **2a** in DCE solvent



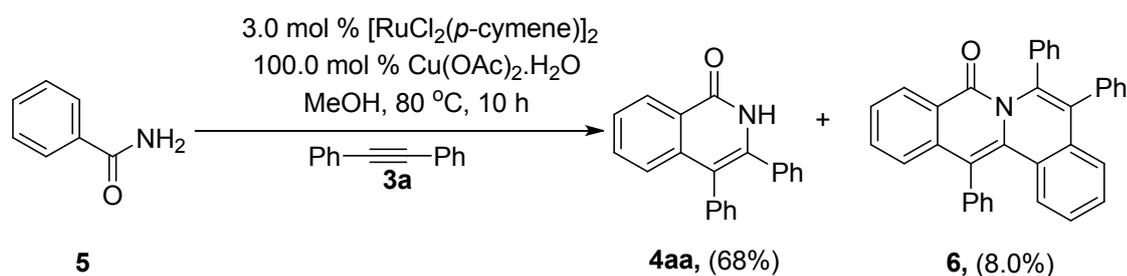
In an flame dried round bottomed flask equipped with a stir bar, we added a mixture of iminophosphorane **2a** (0.39 mmol) (150 mg), in 5.0 mL solvent followed by the addition of alkyne **3a** (0.312 mmol) (56 mg),  $[\text{RuCl}_2(p\text{-cymene})]_2$  (3.0 mol %) and  $\text{Cu}(\text{OAc})_2 \cdot \text{H}_2\text{O}$  (0.39 mmol) (71 mg) at 80 °C. After completion of the reaction in 6 h, monitored by tlc, the resulting mixture was evaporated to remove DCE. After that diluted with  $\text{H}_2\text{O}$  (10 mL) and mixture was extracted with EtOAc (3x20 mL). The combined organic layers were washed

with brine (20 mL) and dried over Na<sub>2</sub>SO<sub>4</sub>. The solvent was evaporated in vacuo and the remaining residue was purified by column chromatography on silica gel (*n*-hexanes/EtOAc 30-40%) to yield product **4aa** (88%, 102 mg).

#### D. Treatment of presynthesized benzamide **5** with unactivated internal alkyne **3a** in

MeOH

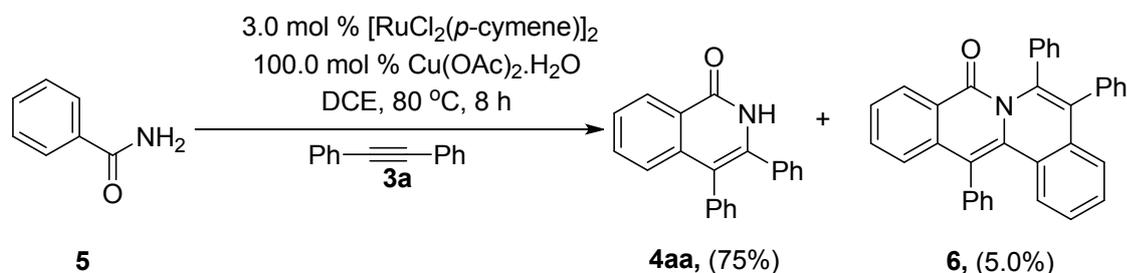
**Scheme 4.** Synthesis of **4aa** from benzamide **5** in MeOH solvent



In a flame dried round bottomed flask equipped with a stir bar, we added a mixture of benzamide **5** (1.23 mmol) (150 mg), in 5.0 mL solvent followed by the addition of alkyne **3a** (0.99 mmol) (176 mg), [RuCl<sub>2</sub>(*p*-cymene)]<sub>2</sub> (3.0 mol%) and Cu(OAc)<sub>2</sub>·H<sub>2</sub>O (1.23 mmol) (223 mg). After completion of the reaction in 10 h, monitored by tlc, the resulting mixture was evaporated to remove MeOH. After that diluted with H<sub>2</sub>O (10 mL) and mixture was extracted with EtOAc (3x20 mL). The combined organic layers were washed with brine (20 mL) and dried over Na<sub>2</sub>SO<sub>4</sub>. The solvent was evaporated in vacuo and the remaining residue was purified by column chromatography on silica gel (*n*-hexanes/EtOAc 5% - 40%) to yield products **4aa** (68%, 249 mg) and **6** (8.0%, 46 mg).

#### E. Treatment of presynthesized benzamide with unactivated internal alkyne **3a** in DCE

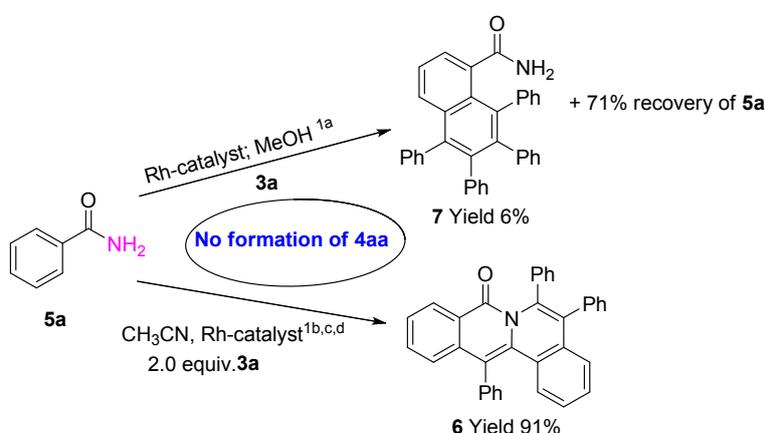
**Scheme 5.** Synthesis of **4aa** from benzamide **5** in DCE solvent



In a flame dried round bottomed flask equipped with a stir bar, we added a mixture of benzamide **5** (1.23 mmol) (150 mg), in 5.0 mL solvent followed by the addition of alkyne **3a**

(0.99 mmol) (176 mg),  $[\text{RuCl}_2(p\text{-cymene})]_2$  (3.0 mol%) and  $\text{Cu}(\text{OAc})_2 \cdot \text{H}_2\text{O}$  (1.23 mmol) (223 mg). After completion of the reaction in 8 h, monitored by tlc, the resulting mixture was evaporated to remove DCE. After that diluted with  $\text{H}_2\text{O}$  (10 mL) and mixture was extracted with EtOAc (3x20 mL). The combined organic layers were washed with brine (20 mL) and dried over  $\text{Na}_2\text{SO}_4$ . The solvent was evaporated in vacuo and the remaining residue was purified by column chromatography on silica gel (*n*-hexanes/EtOAc 5% - 40%) to yield products **4aa** (75%, 276 mg) and **6** (5.0%, 29 mg).

**Scheme 6. Literature reports dealing with transformation of primary benzamide 5a to undesired products 6 and 7<sup>1</sup>**



Literature reports on the transformation of primary benzamide **5** to undesired products **6** and **7** with no formation of **4aa**

Formation of **4aa** from **5a** as depicted in (Scheme 6) was in contrast to literature reports<sup>1</sup> where pre-synthesized **5a** in general has been demonstrated to be resistant towards oxidative annulation affording either **7** or a tricyclic compound **6** as the major product (Scheme 6). Notably, Jaganmohan et al<sup>2</sup> have demonstrated formation of quinolinones from benzonitrile via benzamide with no detectable formation of either **6** or **7**.

## II. <sup>1</sup>H and <sup>31</sup>P NMR Experiments

Experimental evidence for the role and stability of the in situ generated iminophosphoranes during the transformation of **1a** in DCE and to rule out involvement of in situ benzamide was established by <sup>1</sup>H and <sup>31</sup>P experiments. Initially **1a** was allowed to react with **3d** in DCE for 2 h at 80 °C in the presence of (1.0 equiv)  $\text{PPh}_3$ , (3.0 mol %) of  $[\text{Ru}(p\text{-cymene})\text{Cl}_2]_2$ , (1.0 equiv)  $\text{Cu}(\text{OAc})_2 \cdot \text{H}_2\text{O}$  and the reaction mixture after passing through a small celite bed was

evaporated to complete dryness. The  $^1\text{H}$  NMR of the crude product was recorded in  $\text{CDCl}_3$  and compared with presynthesized purified iminophosphorane **2a** and **4ad** (Figure 2)

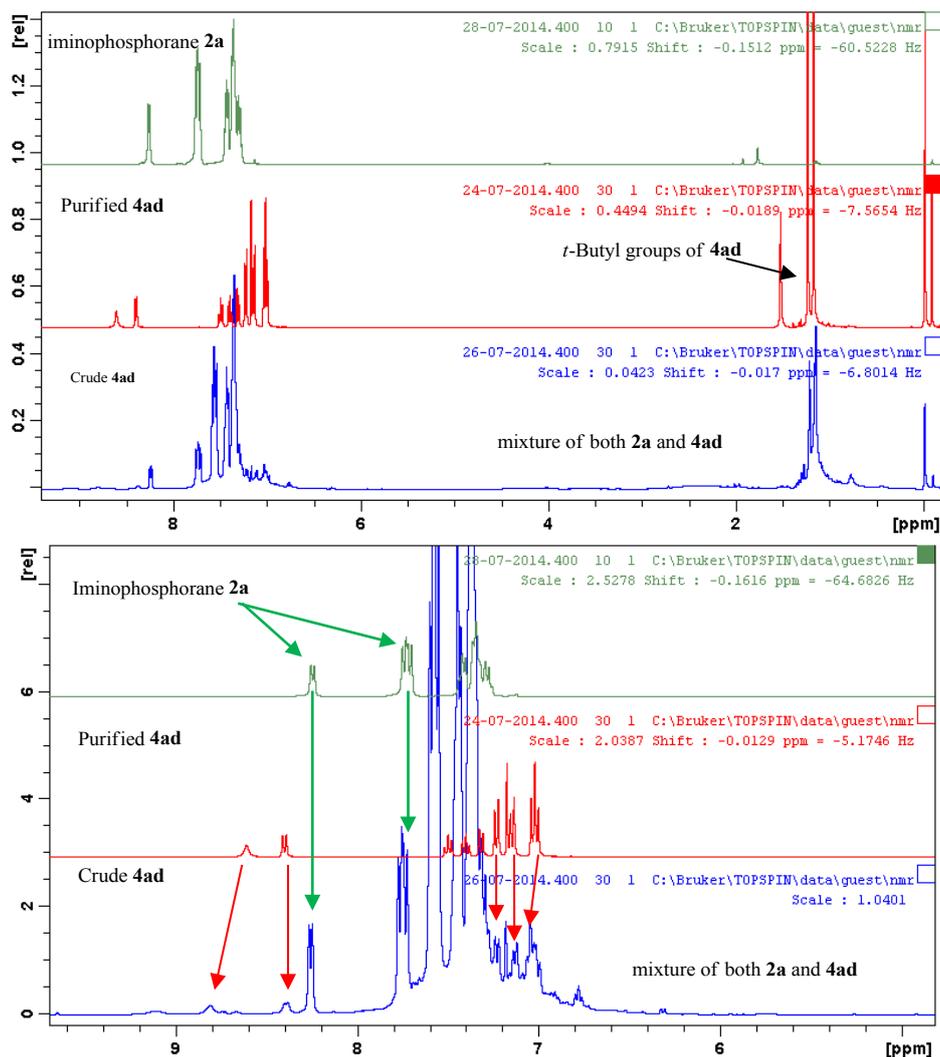


Fig 2: Comparative  $^1\text{H}$  spectra of purified **2a** and **4ad** with crude **4ad**

As is evident, NMR peaks corresponding to the iminophosphorane **2a** and **4ad** can be seen in the crude **4ad**.

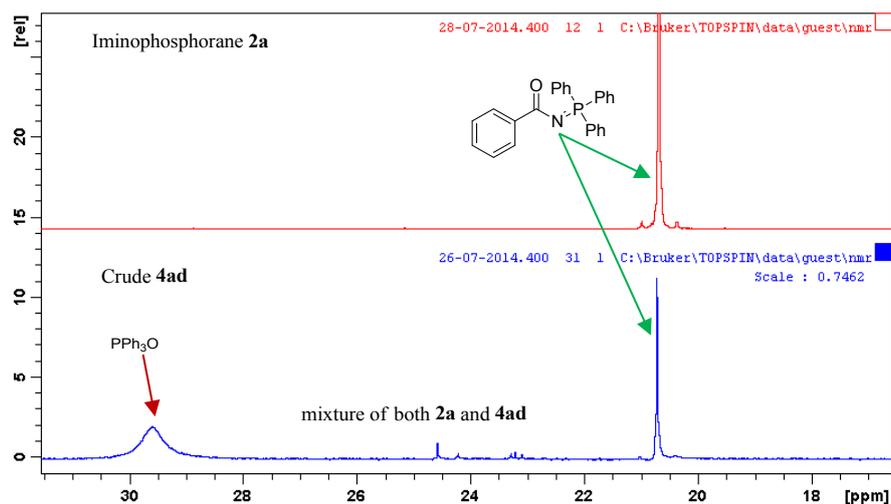
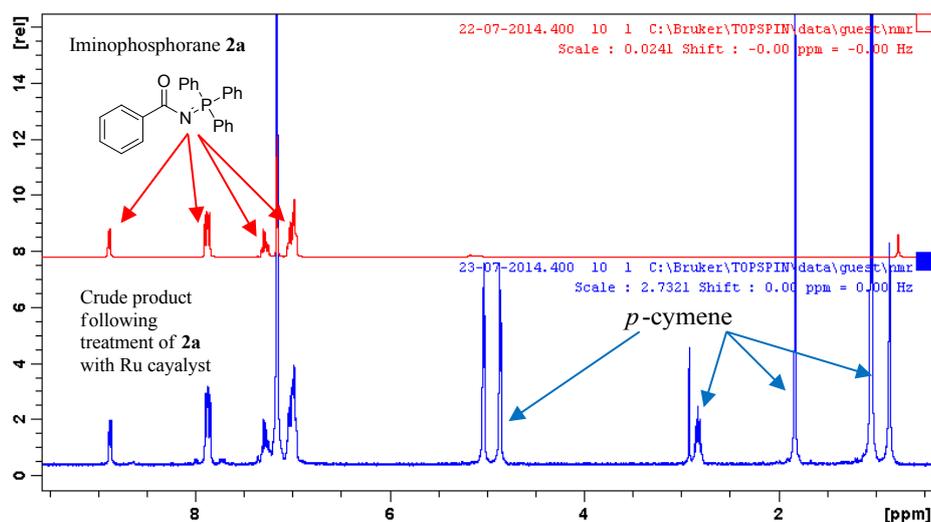
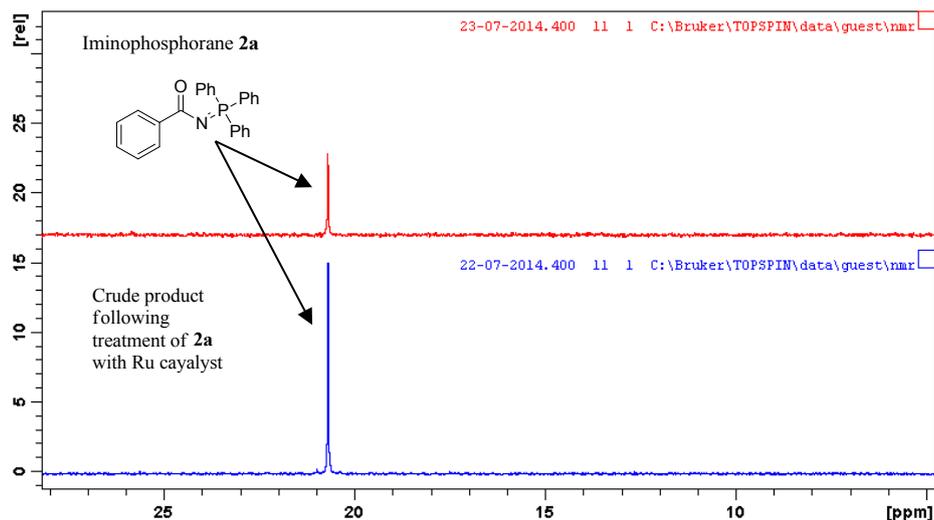


Fig 3:  $^{31}\text{P}$  NMR comparison of **2a** with crude product **4ad**

Next, the  $^{31}\text{P}$  NMR was recorded for the crude reaction mixture and presynthesized iminophosphorane **2a** in  $\text{CDCl}_3$  and a comparative profile has been depicted in Figure 3. As is evident, a peak corresponding to the P present in **2a** at 20.0 ppm can be seen in the crude **4ad** at 20.68 ppm along with formation of triphenyl phosphine oxide ( $\text{PPh}_3\text{O}$ ) as a byproduct at 29.5 ppm. The mass spectrum of the crude reaction mixture also showed the presence of iminophosphorane **2a** corresponding to 382 Da, **4ad** corresponding to 410 Da and  $\text{PPh}_3\text{O}$  corresponding to 279 Da (for mass spectrum see below). The findings suggest that in DCE transformation of **1a** to **4ad** proceeds via in situ formation of iminophosphorane **2a**.

Next in order to check the stability of the N-P bond of **2a**, it was exposed alone (in the absence of **3d**) to  $[\text{Ru}(p\text{-cymene})\text{Cl}_2]_2$  1.0 equiv for 36 h at 80 °C in DCE.<sup>16</sup> After this, the reaction mixture was evaporated to dryness and both  $^1\text{H}$  and  $^{31}\text{P}$  NMR of the crude reaction mixture was recorded in  $\text{C}_6\text{D}_6$  solvent and a comparative profile has been depicted in Figure 4.





**Fig 4:** Comparative  $^1\text{H}$  NMR and  $^{31}\text{P}$  NMR for the stability profile of **2a** with the crude product obtained following treatment of **2a** with Ru-catalyst

As is evident, in situ formation of benzamide **5** was not observed instead  $^1\text{H}$  and  $^{31}\text{P}$  signals can be seen due to the unchanged iminophosphorane **2a** even after 36 h of prolonged heating. The findings yet again rule out involvement of in situ benzamide and confirm involvement of the in situ generated iminophosphorane **2a** as the exclusive intermediate during the transformation of **1a** to **4ad** in DCE.

**III General Information and methods.** All reagents and solvents were purchased from commercial sources and used without purification. NMR spectra were recorded with a 300, 400 MHz spectrometers for  $^1\text{H}$  NMR, 75, 100 MHz for  $^{13}\text{C}$  NMR, and 161.9 MHz for  $^{31}\text{P}$  NMR. Chemical shifts  $\delta$  are given in ppm relative to the residual signals of tetramethylsilane in  $\text{CDCl}_3$  or deuterated solvent  $\text{CDCl}_3/\text{DMSO}-d_6$  and  $\text{C}_6\text{D}_6$  for  $^1\text{H}$  and  $^{13}\text{C}$  NMR. Multiplicities are reported as follows: singlet (s), doublet (d), doublet of doublets (dd), doublet of triplets (dt), triplet (t), quartet (q), multiplet (m), broad singlet (bs). HRMS were obtained using the electro spray ionization (ESI) technique and a time-of-flight (TOF) analyzer. Column chromatography was performed using silica gel (100-200 mesh) as the stationary phase. All reactions were monitored by thin layer chromatography (TLC). The purity and characterization of these compounds were further established using HR/ESI Mass

spectroscopy. Melting points were measured on a capillary melting point apparatus and are uncorrected.

#### **IV: General Procedure for the Preparation of Aryl acyl azide (1a-1n)**

Starting materials **1a-n** were prepared according to the literature<sup>34</sup> procedure (the yields were not optimised). To a solution of acyl chloride (20 mmol) in acetone (20 mL) at 0 °C added drop wise solution of sodium azide (1.99 g, 30 mmol) in water (10 mL) (over 1 h). The reaction mixture was warmed to room temperature and stirred for 8-12 h. Acetone was removed under reduced pressure and the reaction mixture was extracted with EtOAc (30 mL x 3). The combined organic layers were dried over Na<sub>2</sub>SO<sub>4</sub> and solvent was evaporated in vacuo. The residue was purified using silica gel column chromatography (*n*-hexanes/EtOAc 9:1ratio).

Compounds **1a**<sup>3a</sup>, **1b**<sup>3a</sup>, **1f**<sup>3a</sup>, **1h**<sup>3a</sup>, **1j**<sup>3a</sup>, **1k**<sup>4</sup> are reported and their corresponding data matches well with corresponding literature data.

#### **V. General procedure for the ruthenium-catalyzed synthesis of isoquinolone derivatives**

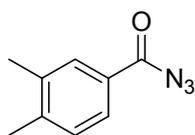
**(4):**

In a flame dried round bottomed flask equipped with a stir bar, we added a mixture of acyl azide **1** (1.02 mmol), in 5.0 mL solvent followed by the addition of Triphenylphosphine (TPP) (1.02 mmol), we then monitored the formation of iminophosphorane by tlc after 30 min of stirring at 80 °C. Next we added alkyne **3** (0.81 mmol), [RuCl<sub>2</sub>(*p*-cymene)]<sub>2</sub> (3.0 mol%) and Cu(OAc)<sub>2</sub>·H<sub>2</sub>O (1.02 mmol). After completion of reaction monitored by tlc, the resulting mixture was diluted with EtOAc, filtered through a pad of celite, and the solvent

was evaporated. At ambient temperature, H<sub>2</sub>O (20 mL) was added and the reaction mixture was extracted with EtOAc (3-20 mL). The combined organic layers were washed with brine (20 mL) and dried over Na<sub>2</sub>SO<sub>4</sub>. The solvent was evaporated in vacuo and the remaining residue was purified by column chromatography on silica gel (*n*-hexanes/EtOAc 3:2 to 4:1) to yield product **4**.

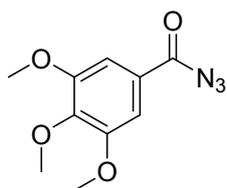
Compounds **4aa**, **4ae**, **4af**, **4ah**, **4ag**, **4ba**, **4fa**, **4ga**, **4ha**, **4ja**, **4ka** and **6** were reported.

**azido(3,4-dimethylphenyl)methanone.(1c)**



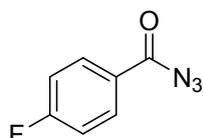
White solid;  $R_f = 0.56$  (10% ethyl acetate/hexane); mp: 48-50 °C; FT-IR (KBr) 3415, 3019, 1669, 1384, 1215 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  7.81 (s, 1H), 7.78-7.76 (m, 1H), 7.22 (d,  $J = 7.9$  Hz, 1H), 2.34 (s, 3H), 2.32 (s, 3H) ppm; <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>):  $\delta$  172.5, 144.1, 137.1, 130.4, 129.9, 128.3, 127.1, 20.1, 19.6 ppm; HRMS (ESI) calcd for C<sub>9</sub>H<sub>10</sub>N<sub>3</sub>O [M + H] 176.0824 found 176.0824.

**azido(3,4,5-trimethoxyphenyl)methanone.(1d)**



White solid;  $R_f = 0.48$  (10% ethyl acetate/hexane); mp: 73-75 °C; FT-IR (KBr) 3406, 3020, 1667, 1466, 1384 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  7.29 (s, 2H), 3.91 (s, 9H) ppm; <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>):  $\delta$  171.8, 153.0, 143.6, 125.5, 106.7, 60.9, 56.3 ppm; HRMS (ESI) calcd for C<sub>10</sub>H<sub>12</sub>N<sub>3</sub>O<sub>4</sub> [M + H] 238.0828 found 238.0836.

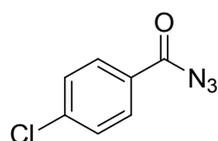
**azido(4-fluorophenyl)methanone.(1e)**



Colorless oil;  $R_f = 0.56$  (10% ethyl acetate/hexane); FT-IR (Neat) 3414, 2181, 1689, 1409, 1216 cm<sup>-1</sup>; <sup>1</sup>H NMR (400 MHz, CDCl<sub>3</sub>):  $\delta$  8.09-8.05 (m, 2H), 7.16-7.12 (m, 2H) ppm; <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>):  $\delta$  171.3, 167.9, 165.4, 132.1 (d,  $J$

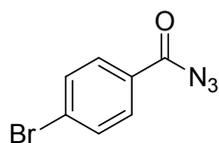
= 10 Hz), 115.9 (d,  $J = 22$  Hz) ppm; HRMS (ESI) calcd for  $C_7H_5N_3OF$  [ $M + H$ ] 166.0417 found 166.0417.

#### azido(4-chlorophenyl)methanone.(1f)



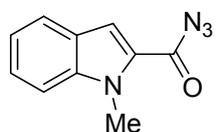
Yellow oil;  $R_f = 0.58$  (10% ethyl acetate/hexane); FT-IR (Neat) 3410, 2401, 1691, 1421, 1280  $cm^{-1}$ ;  $^1H$  NMR (400 MHz,  $CDCl_3$ ):  $\delta$  7.66 (d,  $J = 1.4$  Hz, 1H), 7.61-7.59 (m, 1H), 7.20 (d,  $J = 7.9$  Hz, 1H), 4.86 (s, 2H) ppm;  $^{13}C$  NMR (100 MHz,  $CDCl_3$ ):  $\delta$  171.3, 134.9, 134.2, 132.3, 129.9, 129.4, 127.5 ppm; HRMS (ESI) calcd for  $C_7H_5N_3OCl$  [ $M + H$ ] 182.0121 found 182.0124.

#### azido(4-bromophenyl)methanone.(1g)



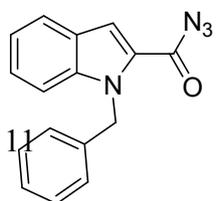
White solid;  $R_f = 0.56$  (10% ethyl acetate/hexane); mp: 58-60 °C; FT-IR (KBr) 3408, 2136, 1686, 1392, 1216  $cm^{-1}$ ;  $^1H$  NMR (400 MHz,  $CDCl_3$ ):  $\delta$  7.90 (d,  $J = 8.7$  Hz, 2H), 7.62 (d,  $J = 8.7$  Hz, 2H) ppm;  $^{13}C$  NMR (100 MHz,  $CDCl_3$ ):  $\delta$  171.6, 132.0, 130.8, 129.7, 129.5 ppm; HRMS (ESI) calcd for  $C_7H_5N_3OBr$  [ $M + H$ ] 225.9616 found 225.9619.

#### azido(1-methyl-1H-indol-2-yl)methanone.(1l)



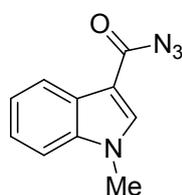
White solid;  $R_f = 0.62$  (10% ethyl acetate/hexane); mp: 62-64 °C; FT-IR (KBr) 3408, 2401, 1677, 1397, 1214  $cm^{-1}$ ;  $^1H$  NMR (400 MHz,  $CDCl_3$ ):  $\delta$  7.64 (d,  $J = 8.0$  Hz, 1H), 7.36-7.31 (m, 3H), 7.15-7.11 (m, 1H), 4.02 (s, 3H) ppm;  $^{13}C$  NMR (100 MHz,  $CDCl_3$ ):  $\delta$  166.4, 140.9, 126.3, 125.8, 123.2, 121.1, 112.7, 110.4, 31.7 ppm; HRMS (ESI) calcd for  $C_{10}H_9N_4O$  [ $M + H$ ] 201.0776 found 201.0785.

#### azido(1-benzyl-1H-indol-2-yl)methanone.(1m)

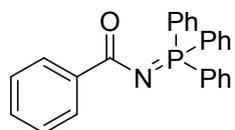


White solid;  $R_f = 0.58$  (10% ethyl acetate/hexane); mp: 68-70 °C; FT-IR (KBr) 3415, 2412, 1682, 1214  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.75 (d,  $J = 8.1$  Hz, 1H), 7.49 (s, 1H), 7.40-7.39 (m, 2H), 7.31-7.27 (m, 3H), 7.25-7.19 (m, 1H), 7.10-7.08 (m, 2H), 5.88 (s, 2H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  166.3, 140.9, 137.9, 128.8, 128.7, 127.4, 126.7, 126.4, 123.3, 121.4, 113.7, 111.0, 48.0 ppm; HRMS (ESI) calcd for  $\text{C}_{16}\text{H}_{13}\text{N}_4\text{O}$  [ $\text{M} + \text{H}$ ] 277.1089 found 277.1089.

**azido(1-methyl-1H-indol-3-yl)methanone.(1n)**



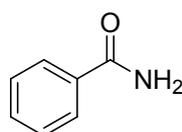
White solid;  $R_f = 0.58$  (10% ethyl acetate/hexane); mp: 84-86 °C; FT-IR (KBr) 3408, 1677, 1214  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  8.28-8.26 (m, 1H), 7.79 (s, 1H), 7.36-7.31 (m, 3H), 3.84 (s, 3H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  167.6, 137.5, 136.4, 126.3, 123.3, 122.6, 121.5, 109.9, 108.2, 33.4 ppm; HRMS (ESI) calcd for  $\text{C}_{10}\text{H}_9\text{N}_4\text{O}$  [ $\text{M} + \text{H}$ ] 201.0776 found 201.0785.



**N-(benzoyl)-triphenyl iminophosphorane (2a)<sup>5</sup>**

White solid,  $R_f = 0.5$  (40% ethyl acetate/hexane); mp 192-194 °C {Lit<sup>5</sup> Rep 196-198 °C}; FT-IR (KBr) 1026, 1112, 1173, 1342, 1437, 1595, 3016  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz), 8.41 (d,  $J = 8.0$  Hz, 2H), 7.91-7.87 (m, 6H), 7.60-7.57 (m, 3H), 7.52-7.42 (m, 9H) ppm;  $^{13}\text{C}$  NMR (100 MHz) 176.3 (d,  $J = 6.0$  Hz), 138.6 (d,  $J = 15.0$  Hz), 133.2 (d,  $J_{\text{C-P}}$ , 7.5 Hz), 132.2 (d,  $J_{\text{C-P}}$ , 1.5 Hz), 13.7, 129.6, 128.9, 128.7, 128.6, 127.9, 127.6 ppm;  $^{31}\text{P}$  NMR (161.9 MHz) 20.68 ppm; HRMS (ESI) calcd for  $\text{C}_{25}\text{H}_{20}\text{NOP}$  [ $\text{M} + \text{H}$ ] 382.1361 Found 381.1363.

**Benzamide.(5a)<sup>6</sup>**



White solid;  $R_f = 0.50$  (50% ethyl acetate/hexane); mp: 108-110 °C {Lit<sup>6</sup> Rep 126-128 °C}; FT-IR (KBr) 3367, 2253, 1673, 1384, 1216  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.83-7.81 (m, 2H), 7.55-7.51 (m, 1H), 7.46-7.43 (m, 2H), 6.22 (brs,

2H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  169.8, 133.43, 131.9, 128.6, 127.4 ppm; HRMS (ESI) calcd for  $\text{C}_7\text{H}_8\text{NO}$  [M + H] 122.0606 found 122.0606.

## VI. Characterisation data of isoquinolin-1(2H)-one (4):

### 3,4-diphenylisoquinolin-1(2H)-one (4aa).<sup>7</sup>

White solid;  $R_f = 0.54$  (40% ethyl acetate/hexane); Yield; 260 mg (86%); mp: 236-238 °C



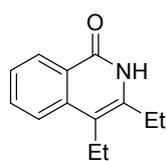
{Lit Rep<sup>7</sup> 242-246 °C }; FT-IR (KBr) 3404, 3018, 1659, 1387, 1215  $\text{cm}^{-1}$ ;

$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.54 (s, 1H), 8.33 (dd,  $J_1 = 7.9$  Hz,  $J_2 = 1.0$  Hz, 1H), 7.89-7.87 (m, 1H), 7.67-7.63 (m, 1H), 7.55-7.51 (m, 1H),

7.47-7.43 (m, 1H), 7.31-7.23 (m, 6H), 7.17-7.15 (m, 3H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  169.6, 162.9, 138.8, 137.2, 135.8, 135.1, 132.8, 132.1, 131.9, 129.4, 128.7, 128.7, 128.5, 128.4, 127.6, 127.5, 127.4, 126.7, 125.8, 125.2, 117.4 ppm; HRMS (ESI) calcd for  $\text{C}_{21}\text{H}_{16}\text{NO}$  [M + H] 298.1232 found 298.1231.

### 3,4-diethylisoquinolin-1(2H)-one (4ae).<sup>7b</sup>

Off-white solid;  $R_f = 0.56$  (40% ethyl acetate/hexane); Yield; 156 mg (76%); mp: 168-170 °C



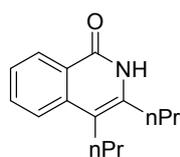
{Lit Rep<sup>7b</sup> 173-175 °C }; FT-IR (KBr) 3399, 3019, 1651, 1384, 1215  $\text{cm}^{-1}$ ;  $^1\text{H}$

NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  11.24 (s, 1H), 8.47 (d,  $J = 7.8$  Hz, 1H), 7.71-7.65 (m, 2H), 7.46-7.42 (m, 1H), 2.79-2.71 (m, 4H), 1.34 (t,  $J = 7.6$  Hz, 3H), 1.21

(t,  $J = 7.5$  Hz, 3H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  164.1, 139.4, 138.4, 132.4, 127.8, 125.3, 122.9, 114.0, 24.3, 19.6, 15.0, 14.1 ppm; HRMS (ESI) calcd for  $\text{C}_{13}\text{H}_{16}\text{NO}$  [M + H] 202.1232 found 202.1238.

### 3,4-dipropylisoquinolin-1(2H)-one (4af).<sup>7a</sup>

Off-white solid;  $R_f = 0.54$  (40% ethyl acetate/hexane); Yield; 182 mg (78%); mp: 174-176 °C



{Lit Rep<sup>7a</sup> 176-180 °C }; FT-IR (KBr) 3399, 3019, 1654, 1384, 1215  $\text{cm}^{-1}$ ;

<sup>1</sup>H NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  10.88 (s, 1H), 8.45 (d,  $J = 8.0$  Hz, 1H), 7.67

(d,  $J = 3.7$  Hz, 1H), 7.46-7.42 (m, 1H), 2.72-2.65 (m, 4H), 1.79-1.70 (m, 3H),

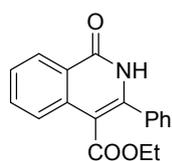
1.65-1.58 (m, 2H), 1.56-1.04 (m, 6H) ppm; <sup>13</sup>C NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  163.9, 138.6,

138.3, 132.4, 127.7, 125.3, 122.2, 123.1, 113.1, 33.0, 28.7, 23.7, 22.9, 14.4, 14.1 ppm;

HRMS (ESI) calcd for  $\text{C}_{15}\text{H}_{20}\text{NO}$  [M + H] 230.1545 found 230.1545.

#### ethyl 1-oxo-3-phenyl-1,2-dihydroisoquinoline-4-carboxylate (4ah).<sup>8</sup>

Off-white solid;  $R_f = 0.50$  (40% ethyl acetate/hexane); Yield; 197 mg (66%); mp: 162-164



°C {Lit Rep<sup>8</sup> 165-167 °C}; FT-IR (KBr) 3389, 1711, 1653, 1489, 1278  $\text{cm}^{-1}$ ;

<sup>1</sup>H NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  10.23 (s, 1H), 8.37 (dd,  $J_1 = 8.0$  Hz,  $J_2 = 0.8$

Hz, 1H), 7.94 (d,  $J = 8.2$  Hz, 1H), 7.75-7.71 (m, 1H), 7.55-7.47 (m, 6H), 4.07

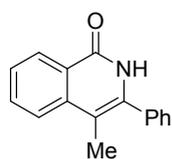
(q,  $J = 14.3$  Hz, 2H), 0.91 (t,  $J = 14.3$  Hz, 3H) ppm; <sup>13</sup>C NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  167.2,

163.1, 141.9, 135.3, 134.8, 133.5, 129.9, 128.8, 128.2, 1277, 127.2, 124.6, 110.4, 61.3, 13.6

ppm; HRMS (ESI) calcd for  $\text{C}_{18}\text{H}_{16}\text{NO}_3$  [M + H] 294.1130 found 294.1130.

#### 4-methyl-3-phenylisoquinolin-1(2H)-one (4ag).<sup>7a</sup>

Off-white solid;  $R_f = 0.48$  (40% ethyl acetate/hexane); Yield; 187 mg (78%); mp: 202-204 °C



{Lit Rep<sup>7a</sup> 200-204 °C }; FT-IR (KBr) 3394, 3019, 1651, 1384, 1215  $\text{cm}^{-1}$ ; <sup>1</sup>H

NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  9.17 (s, 1H), 8.44 (d,  $J = 7.9$  Hz, 1H), 7.64 (d,  $J =$

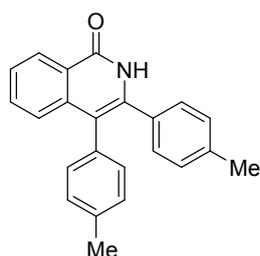
3.6 Hz, 2H), 7.54-7.46 (m, 6H), 2.26 (s, 3H) ppm; <sup>13</sup>C NMR (100 MHz,

$\text{CDCl}_3$ ):  $\delta$  162.8, 138.9, 136.9, 135.4, 132.7, 129.5, 129.1, 128.7, 127.8, 126.4, 125.5, 123.7,

109.2, 13.9 ppm; HRMS (ESI) calcd for  $\text{C}_{16}\text{H}_{14}\text{NO}$  [M + H] 236.1075 found 236.1075.

#### 3,4-bis(4-methylphenyl)isoquinolin-1(2H)-one (4bb).<sup>7b</sup>

Off-white solid;  $R_f = 0.50$  (40% ethyl acetate/hexane); Yield; 282 mg (85%); mp: 250-252



$^{\circ}\text{C}$  {Lit Rep<sup>7b</sup> 254-256  $^{\circ}\text{C}$ }; FT-IR (KBr) 3392, 3019, 1649, 1388,

1215  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.42 (s, 1H), 8.32-8.29

(m, 1H), 7.65-7.60 (m, 1H), 7.52-7.48 (m, 1H), 7.15-7.12 (m, 5H),

7.05-7.03 (m, 3H), 2.29 (s, 3H), 2.25 (s, 3H) ppm;  $^{13}\text{C}$  NMR (100

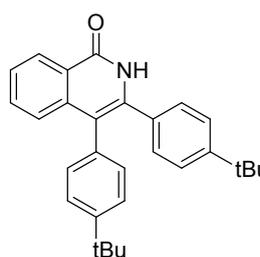
MHz,  $\text{DMSO-}d_6$ ):  $\delta$  162.9, 139.1, 138.6, 137.2, 136.9, 132.9, 132.6, 132.4, 131.7, 129.2,

129.1, 127.5, 126.4, 125.8, 125.1, 117.0, 21.4 ppm; HRMS (ESI) calcd for  $\text{C}_{23}\text{H}_{20}\text{NO}$  [ $\text{M} +$

$\text{H}$ ] 326.1545 found 326.1545.

### 3,4-bis[4-(*tert*-butyl)phenyl]isoquinolin-1(2*H*)-one (4ad).

Off-white solid;  $R_f = 0.52$  (40% ethyl acetate/hexane); Yield; 346 mg (83%); mp: 258-260



$^{\circ}\text{C}$ ; FT-IR (KBr) 3398, 3019, 1651, 1384, 1215  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400

MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.40 (s, 1H), 8.31 (d,  $J = 7.9$  Hz, 1H), 7.64 (d,  $J$

$= 7.0$  Hz, 1H), 7.50 (t,  $J = 8.0$  Hz, 1H), 7.31 (d,  $J = 8.2$  Hz, 2H), 7.23

(d,  $J = 8.4$  Hz, 2H), 7.17-7.13 (m, 3H), 7.07 (d,  $J = 8.2$  Hz, 2H), 1.26

(s, 9H), 1.21 (s, 9H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{DMSO-}d_6 + \text{CDCl}_3$ ):  $\delta$  161.6, 150.2, 149.0,

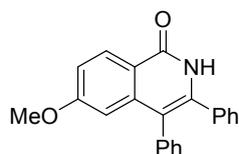
138.2, 138.1, 132.7, 132.0, 131.7, 131.2, 129.3, 126.6, 125.7, 124.8, 124.6, 124.1, 115.2,

34.1, 34.0, 30.9, 30.8 ppm; HRMS (ESI) calcd for  $\text{C}_{29}\text{H}_{32}\text{NO}$  [ $\text{M} + \text{H}$ ] 410.2484 found 410

2484.

### 6-methoxy-3,4-diphenylisoquinolin-1(2*H*)-one (4ba).<sup>7b</sup>

Off-white solid;  $R_f = 0.50$  (40% ethyl acetate/hexane); Yield; 216 mg (78%); mp: 223-225



$^{\circ}\text{C}$  {Lit Rep<sup>7b</sup> 225  $^{\circ}\text{C}$  }; FT-IR (KBr) 3396, 2928, 1644, 1387, 1218  $\text{cm}^{-1}$ ;

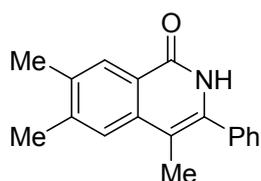
$^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.36 (s, 1H), 8.26 (d,  $J = 8.8$  Hz,

1H), 7.33-7.28 (m, 3H), 7.23 (s, 5H), 7.17-7.14 (m, 3H), 6.52 (d,  $J = 2.4$

Hz, 1H), 3.69 (s, 3H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{DMSO-}d_6 + \text{CDCl}_3$ ):  $\delta$  162.2, 161.8, 140.1, 135.4, 134.4, 131.2, 129.1, 128.8, 127.8, 127.3, 126.6, 118.6, 115.8, 114.4, 106.8, 54.7 ppm; HRMS (ESI) calcd for  $\text{C}_{22}\text{H}_{18}\text{NO}_2$  [M + H] 328.1338 found 328.1338.

#### 4,6,7-trimethyl-3-phenylisoquinolin-1(2H)-one (4bg).

Off-white solid;  $R_f = 0.52$  (40% ethyl acetate/hexane); Yield; 140 mg (62%); mp: 200-202

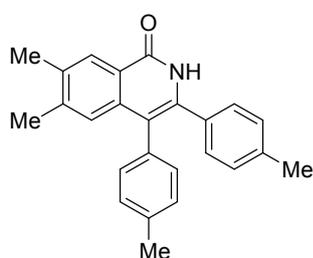


$^{\circ}\text{C}$ ; FT-IR (KBr) 3849, 3394, 1647, 1384, 1215  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  8.51 (s, 1H), 8.24 (s, 1H), 7.52-7.46 (m, 6H), 2.48 (s, 3H), 2.45 (s, 3H), 2.26 (s, 3H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$

162.5, 142.7, 137.1, 135.9, 130.5, 129.3, 128.9, 128.8, 128.4, 127.9, 124.3, 123.5, 109.0, 20.7, 19.8, 13.9 ppm; HRMS (ESI) calcd for  $\text{C}_{18}\text{H}_{18}\text{NO}$  [M + H] 264.1388 found 264.1388.

#### 6,7-dimethyl-3,4-bis(4-methylphenyl)isoquinolin-1(2H)-one (4cb).

Off-white solid;  $R_f = 0.48$  (40% ethyl acetate/hexane); Yield; 212 mg (70%); mp: 272-274

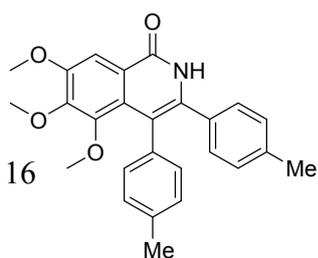


$^{\circ}\text{C}$ ; FT-IR (KBr) 3392, 3019, 1647, 1387, 1215  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.23 (s, 1H), 8.07 (s, 1H), 7.13-7.06 (m, 4H), 7.03-7.00 (m, 4H), 6.91 (s, 1H), 2.37 (s, 3H), 2.30 (s, 3H), 2.24 (s, 3H), 2.22 (s, 3H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{DMSO-}d_6 +$

$\text{CDCl}_3$ ):  $\delta$  162.3, 141.8, 137.6, 137.1, 136.2, 135.3, 133.5, 132.4, 131.8, 129.9, 128.6, 127.2, 125.7, 123.6, 115.5, 21.3, 21.2, 20.5, 19.8 ppm; HRMS (ESI) calcd for  $\text{C}_{25}\text{H}_{24}\text{NO}$  [M + H] 354.1858 found 354.1860.

#### 5,6,7-trimethoxy-3,4-bis(4-methylphenyl)isoquinolin-1(2H)-one (4db).

Red solid;  $R_f = 0.42$  (50% ethyl acetate/hexane); Yield; 178 mg (68%); mp: 258-260  $^{\circ}\text{C}$ ; FT-

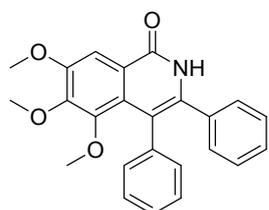


IR (KBr) 3395, 3019, 1645, 1474, 1215  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  8.67 (s, 1H), 7.82 (s, 1H), 7.09-6.99 (m, 8H), 4.03 (s,

3H), 3.91 (s, 3H), 3.15 (s, 3H), 2.32 (s, 3H), 2.29 (s, 3H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  161.7, 153.1, 150.5, 147.9, 138.0, 136.2, 135.6, 135.5, 132.9, 131.1, 129.3, 128.9, 128.8, 127.9, 127.7, 115.0, 104.4, 60.9, 60.8, 56.2, 21.3, 21.3 ppm; HRMS (ESI) calcd for  $\text{C}_{26}\text{H}_{26}\text{NO}_4$  [M + H] 416.1862 found 416.1862.

**5, 6, 7-trimethoxy-3,4-diphenylisoquinolin-1(2H)-one (4da).**

Red solid;  $R_f = 0.44$  (50% ethyl acetate/hexane); Yield; 160 mg (65%); mp: 252-254 °C; FT-

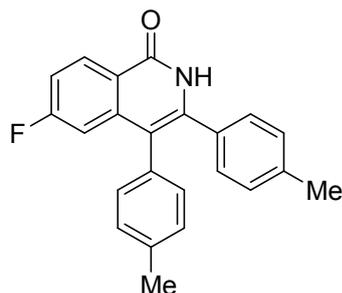


IR (KBr) 3391, 3016, 1642, 1418, 1216  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  8.63 (s, 1H), 7.81 (s, 1H), 7.26 (s, 1H), 7.20-7.13 (m, 9H), 4.02 (s, 3H), 3.90 (s, 3H), 3.13 (s, 3H), ppm;  $^{13}\text{C}$  NMR (100 MHz,

$\text{CDCl}_3$ ):  $\delta$  161.8, 153.2, 150.4, 147.9, 138.7, 136.3, 35.6, 131.4, 129.6, 128.2, 128.1, 126.9, 126.2, 115.3, 104.4, 60.9, 60.8, 56.2 ppm; HRMS (ESI) calcd for  $\text{C}_{24}\text{H}_{22}\text{NO}_4$  [M + H] 388.1549 found 388.1549.

**6-fluoro-3,4-bis(4-methylphenyl)isoquinolin-1(2H)-one (4eb).**

Off-white solid;  $R_f = 0.46$  (40% ethyl acetate/hexane); Yield; 240 mg (77%); mp: 252-254

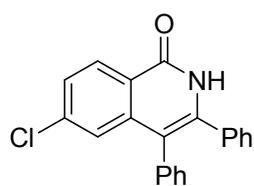


°C; FT-IR (KBr) 3402, 2923, 1651, 1446, 1216  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  8.94 (s, 1H), 8.49-8.45 (m, 1H), 7.19-7.09 (m, 5H), 7.06-6.96 (m, 5H), 2.36 (s, 3H), 2.30 (s, 3H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  166.9, 164.4, 162.5, 141.7 (d,  $J = 10$  Hz), 138.7 (d,  $J = 12$  Hz), 137.2, 132.4, 131.9, 131.5, 130.9 (d,  $J$

= 10 Hz), 129.4, 129.2 (d,  $J = 13$  Hz), 121.7, 116.6, 115.0 (d,  $J = 24$  Hz), 110.9 (d,  $J = 23$  Hz), 21.4 ppm; HRMS (ESI) calcd for  $\text{C}_{23}\text{H}_{19}\text{NOF}$  [M + H] 344.1451 found 344.1451.

**6-chloro-3,4-diphenylisoquinolin-1(2H)-one (4fa).<sup>7b</sup>**

Off-white solid;  $R_f = 0.50$  (40% ethyl acetate/hexane); Yield; 225 mg (82%); mp: 242-244 °C



{Lit Rep<sup>7b</sup> 246-248 °C }; FT-IR (KBr) 3401, 3021, 1652, 1441, 1215

$\text{cm}^{-1}$ ;  $^1\text{H NMR}$  (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.71 (s, 1H), 8.32 (d,  $J = 8.4$

Hz, 1H), 7.57-7.54 (m, 1H), 7.36-7.29 (m, 4H), 7.24 (s, 4H), 7.18-7.16

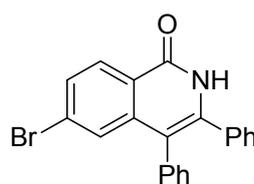
(m, 2H), 7.05 (d,  $J = 2.0$  Hz, 1H) ppm;  $^{13}\text{C NMR}$  (100 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  161.4, 140.3,

139.6, 137.6, 135.1, 134.1, 131.6, 129.7, 129.3, 128.4, 127.7, 127.3, 126.4, 123.7, 123.6,

114.5 ppm; HRMS (ESI) calcd for  $\text{C}_{21}\text{H}_{15}\text{NOCl}$  [ $\text{M} + \text{H}$ ] 332.0842 found 332.0842.

### 6-bromo-3,4-diphenylisoquinolin-1(2H)-one (4ga).<sup>7a</sup>

Off-white solid;  $R_f = 0.46$  (40% ethyl acetate/hexane); Yield; 200 mg (80%); mp: 246-248 °C



{Lit Rep<sup>7a</sup> 250 °C }; FT-IR (KBr) 3410, 2997, 1655, 1389, 1216  $\text{cm}^{-1}$ ;

$^1\text{H NMR}$  (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.71 (s, 1H), 8.24 (d,  $J = 8.5$  Hz,

1H), 7.87 (d,  $J = 8.5$  Hz, 1H), 7.72-7.67 (m, 2H), 7.35-7.31 (m, 3H),

7.91-7.22 (m, 5H), 7.22-7.16 (m, 2H) ppm;  $^{13}\text{C NMR}$  (100 MHz,  $\text{DMSO-}d_6 + \text{CDCl}_3$ ):  $\delta$

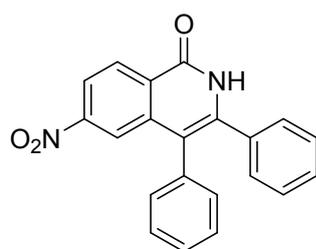
161.2, 140.1, 139.8, 135.0, 134.1, 131.6, 131.5, 129.6, 129.2, 129.0, 128.3, 127.6, 127.3,

126.8, 126.7, 126.7, 123.8, 114.3 ppm; HRMS (ESI) calcd for  $\text{C}_{21}\text{H}_{15}\text{NOBr}$  [ $\text{M} + \text{H}$ ]

376.0337 found 376.0337.

### 6-nitro-3,4-diphenylisoquinolin-1(2H)-one (4ha).<sup>7a</sup>

Off-white solid;  $R_f = 0.42$  (40% ethyl acetate/hexane); Yield; 220 mg (82%); mp 251-252 °C



{Lit Rep<sup>7a</sup> 251-252 °C }; FT-IR (KBr) 3388, 1659, 1531, 1387,

1215  $\text{cm}^{-1}$ ;  $^1\text{H NMR}$  (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  9.99 (s, 1H), 8.58 (d,  $J$

= 8.7 Hz, 1H), 8.22 (s, 2H), 7.36-7.28 (m, 8H), 7.18 (s, 2H) ppm;

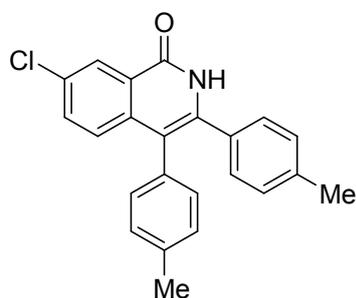
$^{13}\text{C NMR}$  (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  161.8, 150.8, 139.8, 139.7, 134.4,

134.3, 131.7, 129.7, 129.3, 129.0, 128.6, 128.2, 121.3, 120.2, 117.3 ppm; HRMS (ESI) calcd

for  $\text{C}_{21}\text{H}_{15}\text{N}_2\text{O}_3$  [ $\text{M} + \text{H}$ ] 343.1083 found 343.1083.

**7-chloro-3,4-bis(4-methylphenyl)isoquinolin-1(2H)-one (4ib).**

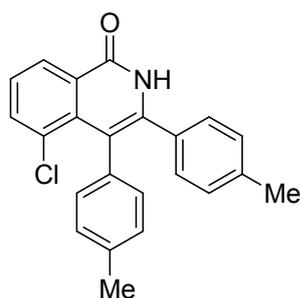
White solid;  $R_f = 0.65$  (40% ethyl acetate/hexane); Yield; (produced in 1:1 ratio); mp: 150-



152 °C; FT-IR (KBr) 3400, 3019, 1632, 1384, 1215  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  9.99 (s, 1H), 8.46 (s, 1H), 7.51 (d,  $J = 8.8$  Hz, 1H), 7.31 (d,  $J = 8.8$  Hz, 1H), 7.14-7.11 (m, 4H), 7.07-7.03 (m, 4H), 2.34 (s, 3H), 2.31 (s, 3H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  162.6, 138.8, 137.6, 133.2, 133.0, 132.6,

132.4, 131.6, 131.5, 129.7, 129.4, 129.3, 129.1, 127.6, 126.8, 117.2, 21.4 ppm; HRMS (ESI) calcd for  $\text{C}_{23}\text{H}_{19}\text{NOCl}$  [ $\text{M} + \text{H}$ ] 360.1155 found 360.1157.

**5-chloro-3,4-bis(4-methylphenyl)isoquinolin-1(2H)-one (4ib').**

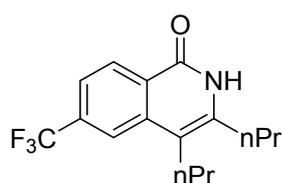


White solid;  $R_f = 0.56$  (50% ethyl acetate/hexane); mp: 200-202 °C; FT-IR (KBr) 3400, 3019, 1632, 1384, 1215  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  9.30 (s, 1H), 8.45 (d,  $J = 7.9$  Hz, 1H), 7.64-7.62 (m, 1H), 7.39-7.35 (m, 1H), 7.02-6.98 (m, 8H), 2.31 (s, 3H), 2.28 (s, 3H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  162.1, 139.7, 138.4,

136.6, 135.1, 134.3, 132.5, 132.1, 131.3, 129.3, 129.2, 128.8, 128.2, 127.2, 126.8, 115.6, 21.4, 21.3 ppm; HRMS (ESI) calcd for  $\text{C}_{23}\text{H}_{19}\text{NOCl}$  [ $\text{M} + \text{H}$ ] 360.1155 found 360.1157.

**3,4-dipropyl-6-(trifluoromethyl)isoquinolin-1(2H)-one (4cf).**

Off-white solid;  $R_f = 0.48$  (40% ethyl acetate/hexane); Yield; 149 mg (72%); mp: 120-122



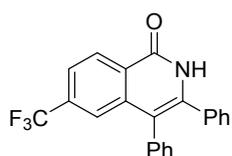
°C; FT-IR (KBr) 3430, 1650, 1384, 1217  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  11.27 (s, 1H), 8.55 (d,  $J = 8.3$  Hz, 1H), 7.92 (s, 1H), 7.64 (d,  $J = 8.3$ , 1H), 7.02 (s, 1H), 2.75-2.69 (m, 4H), 1.82-1.72 (m, 2H),

1.64-1.57 (m, 2H), 1.09-1.01 (m, 6H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ):  $\delta$  163.3, 140.2,

138.6, 134.0 (d,  $J = 29.0$  Hz), 128.8, 127.2, 125.5, 122.8, 121.4, 120.5, 113.1, 33.1, 28.5, 23.7, 22.9, 14.2 (d,  $J = 21.0$  Hz) ppm; HRMS (ESI) calcd for  $C_{16}H_{19}NOF_3$  [M + H] 298.1419 found 298.1419.

### 3,4-diphenyl-6-(trifluoromethyl)isoquinolin-1(2H)-one (4ja).<sup>7a,7b</sup>

Off-white solid;  $R_f = 0.62$  (50% ethyl acetate/hexane); Yield; 224 mg (88%); mp 228-230 °C



{Lit Rep<sup>7a</sup> 225 °C }; FT-IR (KBr) 3390, 3019, 1660, 1382, 1223  $cm^{-1}$ ;  $^1H$

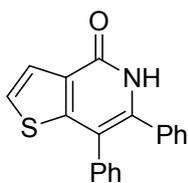
NMR (400 MHz,  $CDCl_3$ ):  $\delta$  9.01 (s, 1H), 8.59 (d,  $J = 8.0$  Hz, 1H), 7.71-

7.69 (m, 1H), 7.62 (s, 1H), 7.35 -7.34 (m, 3H), 7.3-7.26 (m, 3H), 7.24 –

7.21 (m, 2H), 7.18-7.16 (m, 2H) ppm;  $^{13}C$  NMR (100 MHz,  $CDCl_3$ ):  $\delta$  162.5, 138.9 (d,  $J = 5.0$  Hz), 134.74 (d,  $J = 25.0$  Hz), 131.8, 129.4, 129.1, 128.8 (d,  $J = 7.0$  Hz), 128.5, 127.7, 127.3, 122.9, 122.7, 117.1 ppm; HRMS (ESI) calcd for  $C_{22}H_{15}NOF_3$  [M + H] 366.1106 found 366.1106.

### 4,5-Diphenylthieno[2,3-c]pyridin-7(6H)-one (4ka).<sup>7b</sup>

White solid;  $R_f = 0.56$  (50% ethyl acetate/hexane); Yield; 179 mg (60%); mp 263-265 °C;



{Lit Rep<sup>7b</sup> 265-267 °C } FT-IR (KBr) 3390, 3019, 1660, 1384, 1216  $cm^{-1}$ ;  $^1H$

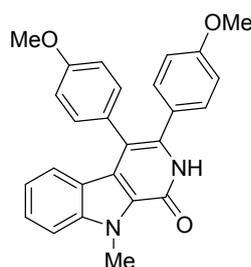
NMR (400 MHz,  $DMSO-d_6$ ):  $\delta$  11.76 (s, 1H), 8.03 (d,  $J = 5.2$  Hz, 1H), 7.28-

7.23 (m, 8H), 7.16-7.14 (m, 2H), 6.93 (d,  $J = 5.2$  Hz, 1H) ppm;  $^{13}C$  NMR

(100 MHz,  $DMSO-d_6$ ):  $\delta$  158.0, 146.7, 139.8, 136.3, 134.1, 133.9, 130.7, 130.0, 128.3, 128.2, 128.0, 127.8, 126.9, 124.5, 114.7 ppm; HRMS (ESI) calcd for  $C_{19}H_{14}NOS$  [M + H] 304.0796 found 304.0796.

### 3,4-bis(4-methoxyphenyl)-9-methyl-2,9-dihydro-1H- $\beta$ -carbolin-1-one (4lc).

Light-brown solid;  $R_f = 0.30$  (50% ethyl acetate/hexane); Yield; 199 mg (65%); mp: 271-273

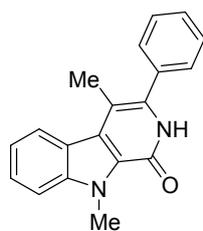


°C; FT-IR (KBr) 3421, 3019, 1640, 1384, 1215  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.48 (s, 1H), 7.62 (d,  $J = 8.4$  Hz, 1H), 7.42-7.39 (m, 1H), 7.18-7.16 (m, 4H), 6.96-6.93 (m, 3H), 6.79 (d,  $J = 8.8$  Hz, 1H), 6.67 (d,  $J = 8.1$  Hz, 1H), 4.31 (s, 3H), 3.79 (s, 3H), 3.72 (s, 3H)

ppm;  $^{13}\text{C}$  NMR of this compound was not obtained due to its very very low solubility; HRMS (ESI) calcd for  $\text{C}_{26}\text{H}_{23}\text{N}_2\text{O}_3$  [ $\text{M} + \text{H}$ ] 411.1709 found 411.1709.

#### 4,9-dimethyl-3-phenyl-2,9-dihydro-1H- $\beta$ -carboline-1-one (4lg).

Orange solid;  $R_f = 0.50$  (50% ethyl acetate/hexane); Yield; 114 mg (58%); mp: 268-270 °C;

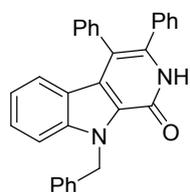


FT-IR (KBr) 3420, 1644, 1215  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.36 (s, 1H), 8.18 (d,  $J = 7.9$  Hz, 1H), 7.68 (d,  $J = 8.4$  Hz, 1H), 7.55-7.46 (m, 6H), 7.26(t,  $J = 7.3$  Hz, 1H), 4.30 (s, 3H), 2.45 (s, 3H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{DMSO-}d_6$ ): 155.6, 140.6, 134.2, 133.9, 129.9, 128.6, 128.3,

126.7, 126.0, 125.8, 122.9, 122.0, 120.4, 110.0, 107.9, 31.5, 15.6 ppm; HRMS (ESI) calcd for  $\text{C}_{19}\text{H}_{19}\text{N}_2\text{O}$  [ $\text{M} + \text{H}$ ] 289.1341 found 289.1340.

#### 9-benzyl-3,4-diphenyl-2,9-dihydro-1H- $\beta$ -carboline -1-one (4ma).

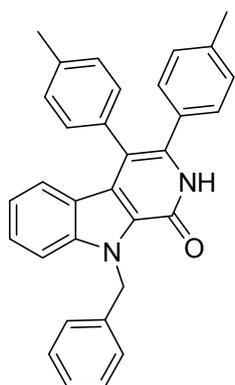
Light- Yellow solid;  $R_f = 0.60$  (50% ethyl acetate/hexane); Yield; 157 mg (68%); mp: 298-



300 °C; FT-IR (KBr) 3427, 2989, 1644, 1386, 1219  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.73 (s, 1H), 7.64 (d,  $J = 8.5$  Hz, 1H), 7.38-7.36 (m, 3H), 7.32-7.28 (m, 8H), 7.28-7.23 (m, 4H), 6.88 (t,  $J = 7.9$  Hz, 1H), 6.58 (d,

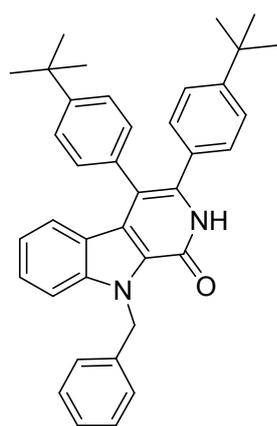
$J = 8.2$  Hz, 1H), 6.18 (s, 2H) ppm;  $^{13}\text{C}$  NMR of this compound was not obtained due to its very very low solubility; HRMS (ESI) calcd for  $\text{C}_{30}\text{H}_{23}\text{N}_2\text{O}$  [ $\text{M} + \text{H}$ ] 427.1810 found 427.1810.

**9-benzyl-3,4-bis(4-methylphenyl)-2,9-dihydro-1H-pyrido[3,4-b]indol-1-one (4mb)**



Brown solid;  $R_f = 0.48$  (50% ethyl acetate/hexane); Yield; 167 mg (68%); mp: 188-190 °C; FT-IR (KBr) 3402, 2926, 1389  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.62 (s, 1H), 7.62 (d,  $J = 8.0$  Hz, 1H), 7.35-7.33 (m, 1H), 7.30-7.29 (m, 4H), 7.24-7.21 (m, 2H), 7.18-7.15 (m, 5H), 7.06-7.04 (m, 2H), 6.88 (t,  $J = 8.0$  Hz, 1H), 6.62 (d,  $J = 8.0$  Hz, 1H) 6.17 (s, 2H), 2.36 (s, 3H), 2.25 (s, 3H) ppm;  $^{13}\text{C}$  NMR of this compound was

not obtained due to its very very low solubility; HRMS (ESI) calcd for  $\text{C}_{32}\text{H}_{27}\text{N}_2\text{O}$  [M + H] 455.2123 found 455.2130.

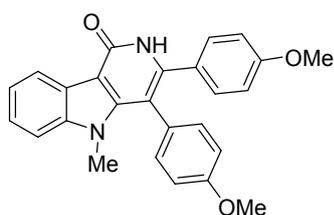


**9-benzyl-3,4-bis(4-(tert-butyl)phenyl)-2,9-dihydro-1H-pyrido[3,4-b]indol-1-one (4md)**

Brown solid;  $R_f = 0.55$  (50% ethyl acetate/hexane); Yield; 210 mg (72%); mp 278-280 °C; FT-IR (KBr) 3401, 3020, 1639  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.58 (s, 1H), 7.62 (d,  $J = 8.0$  Hz, 1H), 7.39-7.38 (m, 2H), 7.30-7.29 (m, 5H), 7.23-7.20 (m, 7H), 6.85 (t,  $J = 8.0$  Hz, 1H), 6.52 (d,  $J = 8.0$  Hz, 1H) 6.16 (s, 2H), 1.31 (s, 9H), 1.23 (s, 9H) ppm;  $^{13}\text{C}$  NMR of this compound was not obtained due to its very very low solubility; HRMS (ESI) calcd for  $\text{C}_{38}\text{H}_{39}\text{N}_2\text{O}$  [M + H] 539.3062 found 539.3062.

**5-methyl-3,4-bis[4-methoxyphenyl]-2,5-dihydro-1H-pyrido[4,3-b]indol-1-one (4nc).**

Brown solid;  $R_f = 0.30$  (50% ethyl acetate/hexane); Yield; 230 mg (72%); mp: 268-270 °C;

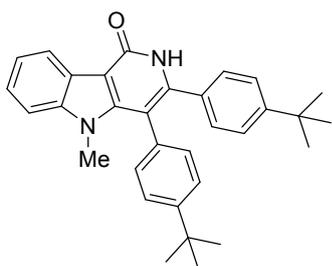


FT-IR (KBr) 3426, 2992, 1634, 1387, 1218  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.30 (s, 1H), 8.26 (d,  $J = 7.2$  Hz, 1H), 7.49 (d,  $J = 8.2$  Hz, 1H), 7.38-7.34 (m, 1H), 7.29-7.25 (m, 1H), 7.22-

7.16 (m, 1H), 6.89 (d,  $J = 8.7$  Hz, 2H), 6.79 (d,  $J = 8.8$  Hz, 1H), 3.76 (s, 3H), 3.72 (s, 3H), 3.16 (s, 3H) ppm;  $^{13}\text{C}$  NMR of this compound was not obtained due to its very very low solubility; HRMS (ESI) calcd for  $\text{C}_{27}\text{H}_{26}\text{NO}_3$  [ $\text{M} + \text{H}$ ] 411.1709 found 411.1709.

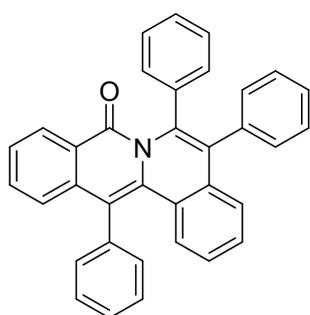
**5-methyl-3,4-bis[4-(*tert*-butyl)phenyl]-2,5-dihydro-1*H*-pyrido[4,3-*b*]indol-1-one (4nd).**

Light-brown solid;  $R_f = 0.48$  (50% ethyl acetate/hexane); Yield; 200 mg (58%); mp  $>300$  °C;



FT-IR (KBr) 3421, 2993, 1646, 1387, 1218  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR (400 MHz,  $\text{DMSO-}d_6$ ):  $\delta$  11.33 (s, 1H), 8.26 (d,  $J = 7.6$  Hz, 1H), 7.49 (d,  $J = 8.2$  Hz, 1H), 7.38-7.34 (m, 1H), 7.31-7.26 (m, 3H), 7.23-7.19 (m, 4H), 7.14 (d,  $J = 8.4$  Hz, 2H), 3.17 (s, 3H), 1.26 (s, 9H),

1.21 (s, 9H) ppm;  $^{13}\text{C}$  NMR of this compound was not due to its very very low solubility; HRMS (ESI) calcd for  $\text{C}_{32}\text{H}_{35}\text{N}_2\text{O}$  [ $\text{M} + \text{H}$ ] 463.2749 found 463.2747.



**5,6,13-triphenyl-13, 13a-dihydro-8*H*-isoquinolino[3,2-*a*]isoquinolin-8-one (6)<sup>9</sup>**

Light-Yellow solid;  $R_f = 0.65$  (20% ethyl acetate/hexane);  $^1\text{H}$  NMR (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  8.25-8.23 (m, 1H), 7.61-7.59 (m, 1H), 7.58-7.52 (m, 5H),

7.50-7.40 (m, 1H), 7.35-7.33 (m, 1H), 7.28-7.25 (m, 4H), 7.24-7.23 (m, 3H), 7.22-7.14 (m, 2H), 7.12-7.01 (m, 4H), 6.89-6.85 (m, 1H) ppm;  $^{13}\text{C}$  NMR (100 MHz,  $\text{CDCl}_3$ ): 162.2, 138.6, 137.1, 136.3, 136.2, 133.8, 133.1, 132.3, 132.2, 131.5, 129.1, 129.0, 128.5, 128.1, 128.0, 127.6, 127.5, 127.1, 126.8, 126.7, 126.5, 126.3, 125.8, 125.7, 125.6, 117.0 ppm; HRMS (ESI) calcd for  $\text{C}_{35}\text{H}_{24}\text{NO}$  [ $\text{M} + \text{H}$ ] 474.1858 found 474.1863.

**Triphenylphosphine Oxide**

White solid;  $R_f = 0.35$  (50% ethyl acetate/hexane);  $^1\text{H NMR}$  (400 MHz,  $\text{CDCl}_3$ ):  $\delta$  7.71-7.60 (m, 6H), 7.56-7.54 (m, 3H), 7.50-7.45 (m, 6H) ppm;  $^{13}\text{C NMR}$  (100 MHz,  $\text{CDCl}_3$ ): 133.0, 132.1, 132.0, 131.9, 128.5, 128.4 ppm

## VII. Spectral Copies of Starting and Final Compounds

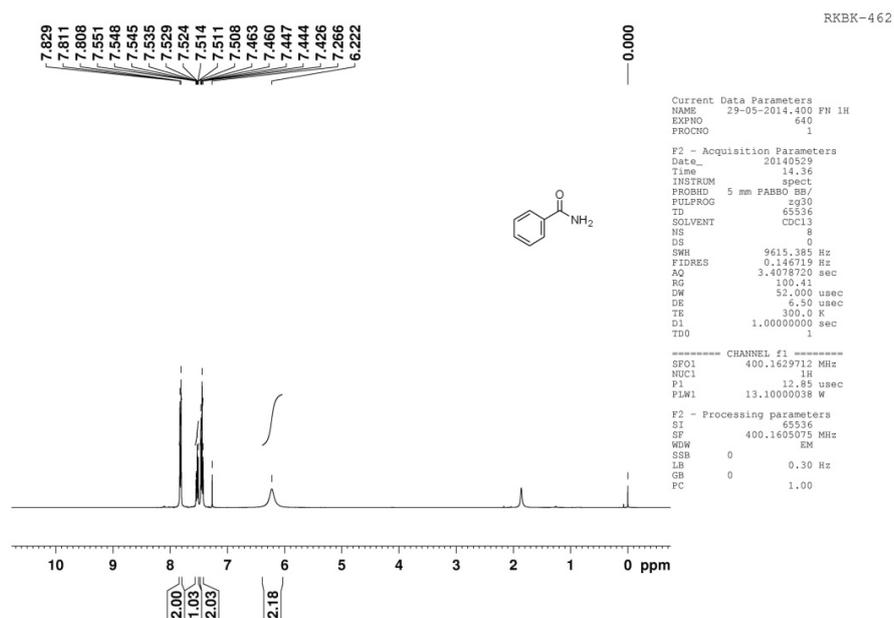
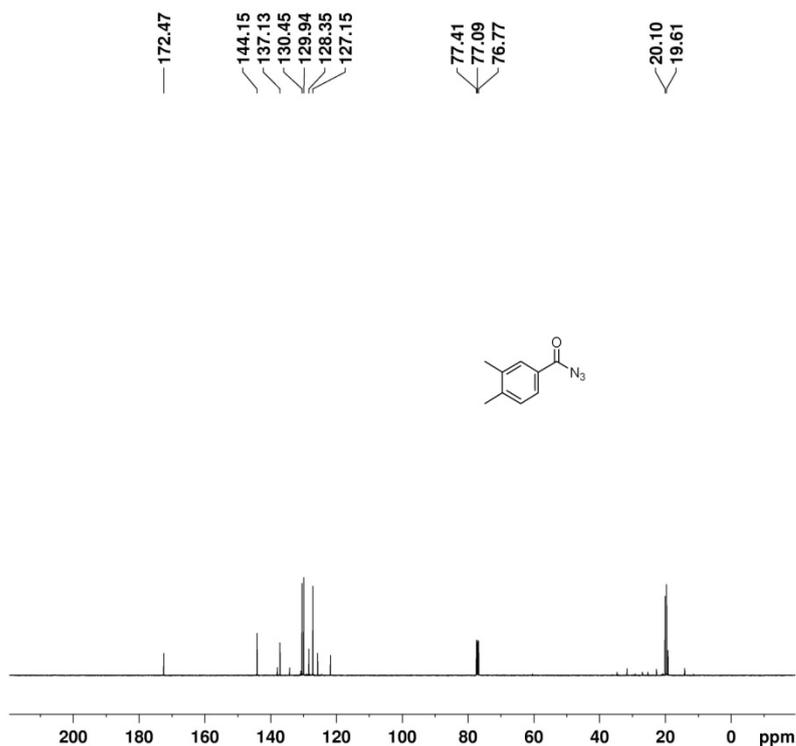


Fig. 5:  $^1\text{H NMR}$  of 5



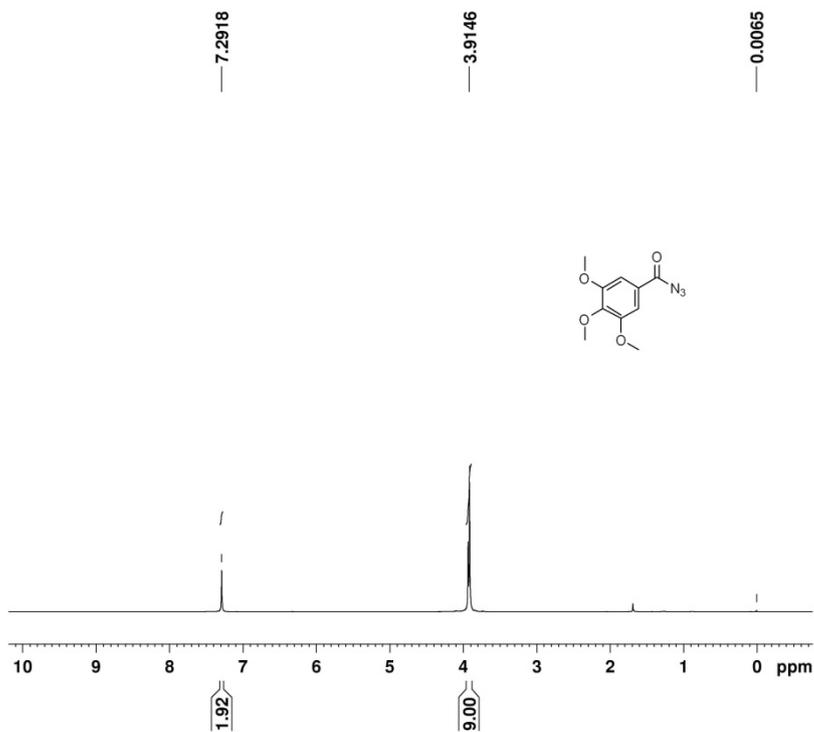


RKKB-466

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FIDRES 0.166708 Hz
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RG 251.08
DW 20.800 usec
DE 6.50 usec
TE 300.2 K
D1 2.0000000 sec
d11 0.3300000 sec
TD0 1
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NUC1 13
P1 1.00 usec
PL1 0.0000000 W
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SFO2 400.1621006 MHz
NUC2 13
P2 1.00 usec
PL2 0.0000000 W
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GB 0
PC 1.40
  
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Fig. 8: <sup>13</sup>C NMR of 1c

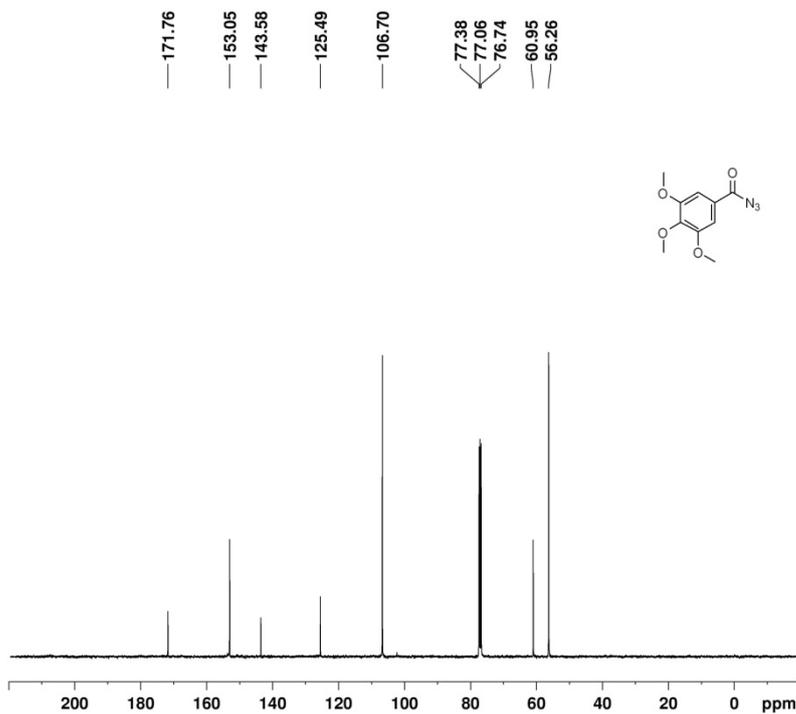


RKKB-A33

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FIDRES 0.144713 Hz
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RG 65.63
DW 32.800 usec
DE 6.50 usec
TE 300.2 K
D1 1.0000000 sec
TD0 1
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NUC1 13
P1 1.00 usec
PL1 13.1000000 W
F2 - Processing parameters
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WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.00
  
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Fig. 9: <sup>1</sup>H NMR of 1d



RKBK-A33

```

Current Data Parameters
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PROCNO    1

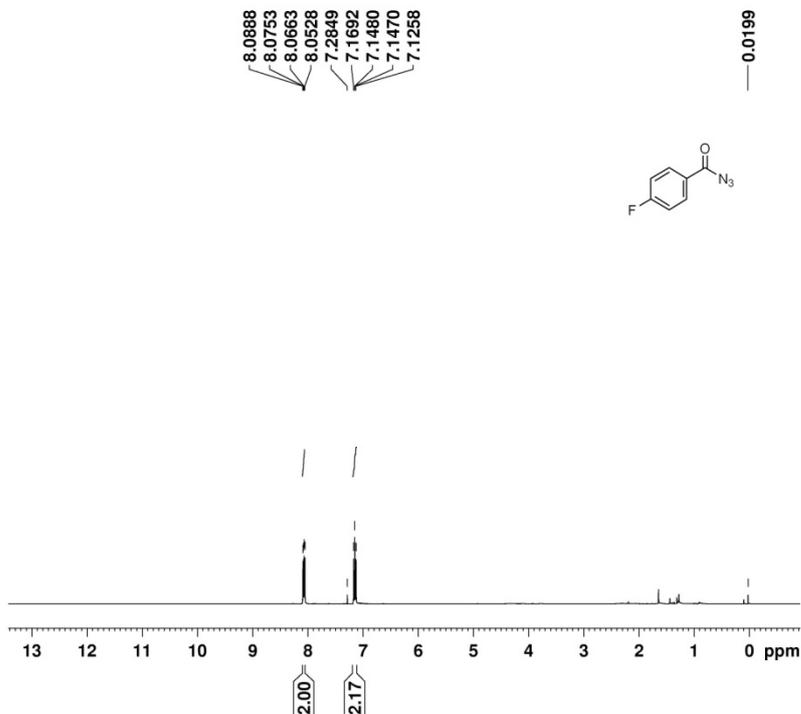
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Date_     20140606
Time      2.21
INSTRUM   spect
PROBHD    5 mm PABBO MQ
PULPROG   zg30
TD         65536
SOLVENT   CDCl3
NS         8
DS         2
SWH        8012.820 Hz
FIDRES     0.122266 Hz
AQ         4.0394465 sec
RG         80.54
DW         62.400 usec
DE         6.50 usec
TE         300.0 K
D1         1.00000000 sec
TDO        1

===== CHANNEL f1 =====
SFO1      100.626120 MHz
NUC1       13C
P1         1.50 usec
PLW1       61.0999967 W

===== CHANNEL f2 =====
SFO2      400.142100 MHz
NUC2       1H
P2         12.85 usec
PLW2       13.10000038 W

F2 - Processing parameters
SI         65536
SF         400.1605000 MHz
WDW        EM
SSB        0
LB         0.30 Hz
GB         0
PC         1.00
  
```

Fig. 10: <sup>13</sup>C NMR of 1d



RKBK-467

```

Current Data Parameters
NAME      05-06-2014.400.1H
EXPNO     610
PROCNO    1

F2 - Acquisition Parameters
Date_     20140606
Time      2.21
INSTRUM   spect
PROBHD    5 mm PABBO BBI/
PULPROG   zg30
TD         65536
SOLVENT   CDCl3
NS         8
DS         2
SWH        8012.820 Hz
FIDRES     0.122266 Hz
AQ         4.0394465 sec
RG         80.54
DW         62.400 usec
DE         6.50 usec
TE         300.0 K
D1         1.00000000 sec
TDO        1

===== CHANNEL f1 =====
SFO1      400.1629712 MHz
NUC1       1H
P1         12.85 usec
PLW1       13.10000038 W

F2 - Processing parameters
SI         65536
SF         400.1605000 MHz
WDW        EM
SSB        0
LB         0.30 Hz
GB         0
PC         1.00
  
```

Fig. 11: <sup>1</sup>H NMR of 1e

RKBK-467

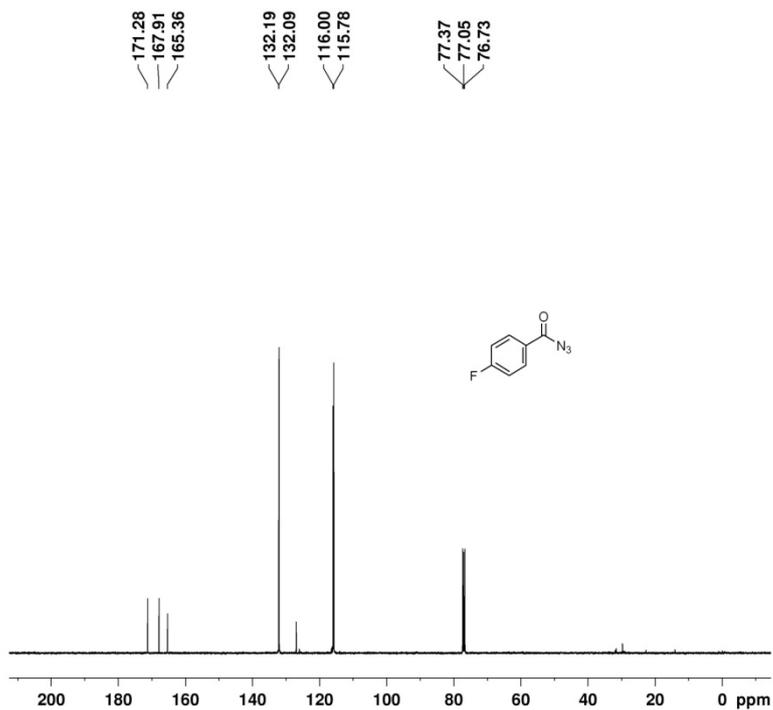


Fig. 12: <sup>13</sup>C NMR of 1e

RKBK-468

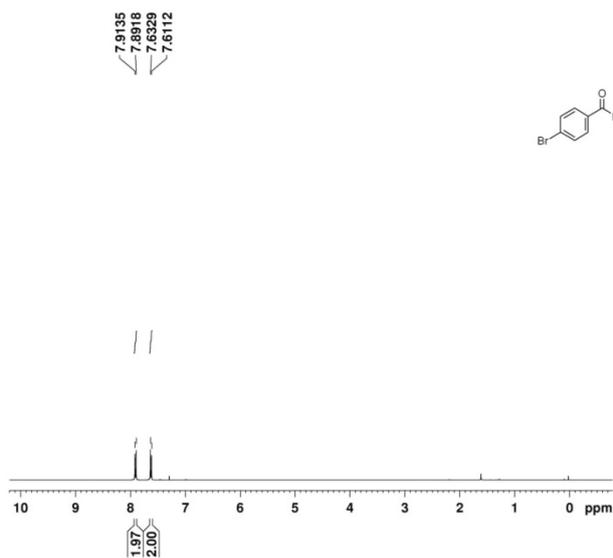
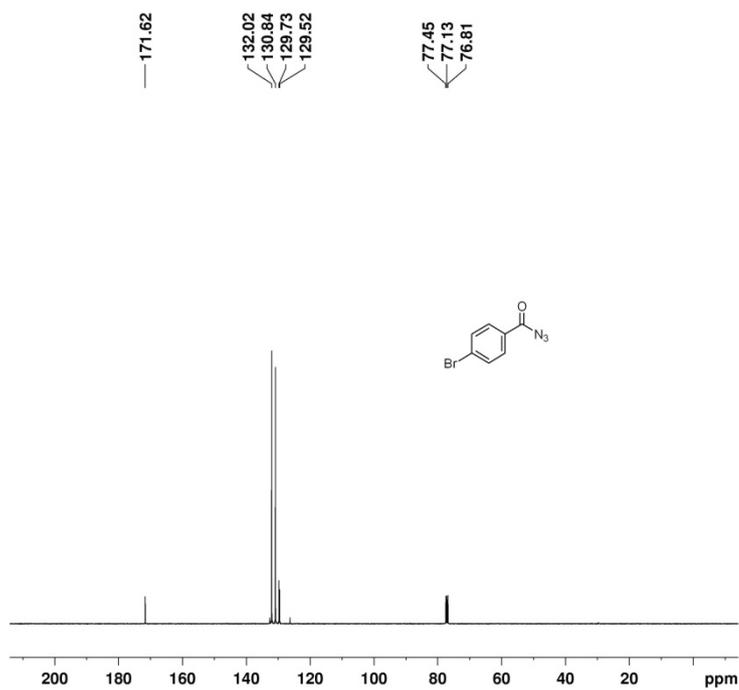


Fig. 13: <sup>1</sup>H NMR of 1f

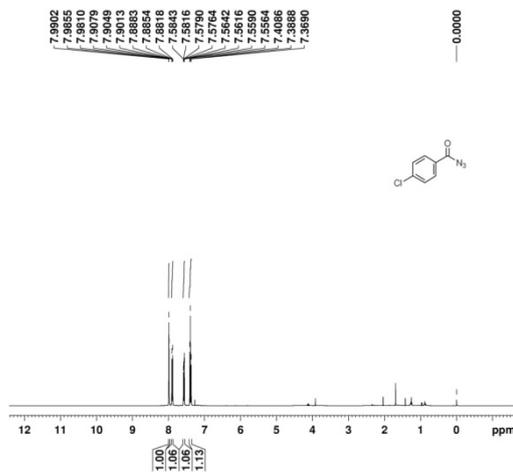


RKKB-468

```

Name: 4-Bromoacetophenone
Date: 05-06-2014 09:04 AM, 13C
P1: 100.625000 MHz
F2 - Acquisition Parameters
Date_: 05-06-2014
Time: 09:04:00
INSTRUM: spect
PROBHD: 5 mm BBO-1H/1
PULPROG: zgpg30
AQ: 0.16300000
RG: 655.36
SFO: 125.761
FIDRES: 0.00010000
AQRES: 0.00010000
DELTA: 0.00000000
DELTAT: 0.00000000
TE: 300.2
D1: 1.00000000
d11: 0.00000000
d12: 0.00000000
d13: 0.00000000
d14: 0.00000000
===== CHANNEL f1 =====
NUC1: 13C
P1: 0.12000000
PL1: 0.00
PL2: 0.00
PL3: 0.00
PL4: 0.00
===== CHANNEL f2 =====
NUC2: 1H
P1: 0.12000000
PL1: 0.00
PL2: 0.00
PL3: 0.00
PL4: 0.00
===== CHANNEL f3 =====
NUC3: 13C
P1: 0.12000000
PL1: 0.00
PL2: 0.00
PL3: 0.00
PL4: 0.00
F2 - Processing parameters
SI: 32768
SF: 101.6250000 MHz
WDW: EM
SSB: 0
GB: 0
PC: 1.40
  
```

Fig. 14: <sup>13</sup>C NMR of 1f

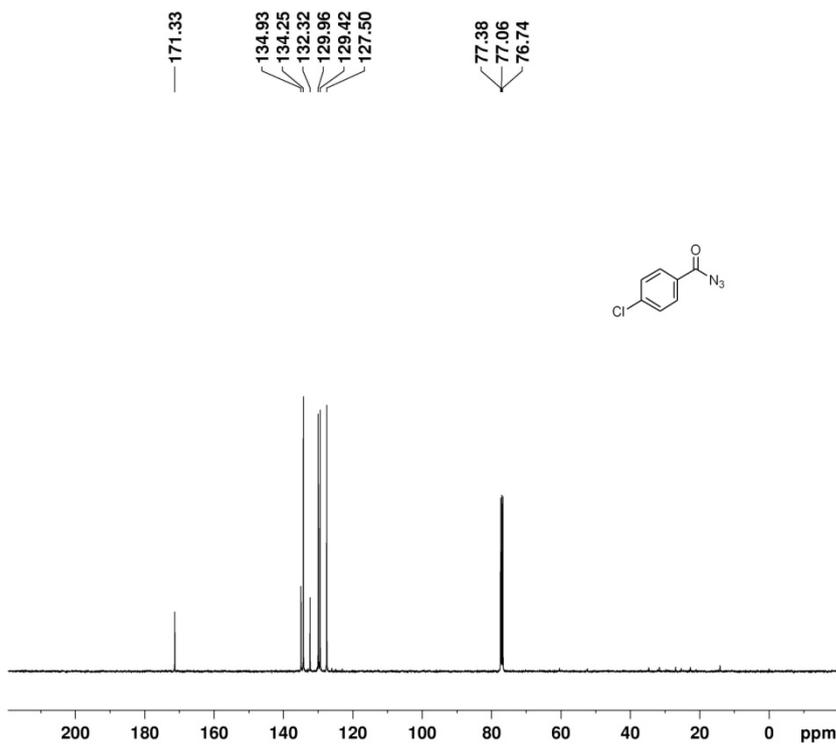


RKKB-465

```

Name: 4-Chloroacetophenone
Date: 05-06-2014 09:04 AM, 1H
P1: 100.625000 MHz
F2 - Acquisition Parameters
Date_: 05-06-2014
Time: 09:04:00
INSTRUM: spect
PROBHD: 5 mm BBO-1H/1
PULPROG: zgpg30
AQ: 0.16300000
RG: 655.36
SFO: 125.761
FIDRES: 0.00010000
AQRES: 0.00010000
DELTA: 0.00000000
DELTAT: 0.00000000
TE: 300.2
D1: 1.00000000
d11: 0.00000000
d12: 0.00000000
d13: 0.00000000
d14: 0.00000000
===== CHANNEL f1 =====
NUC1: 1H
P1: 0.12000000
PL1: 0.00
PL2: 0.00
PL3: 0.00
PL4: 0.00
===== CHANNEL f2 =====
NUC2: 13C
P1: 0.12000000
PL1: 0.00
PL2: 0.00
PL3: 0.00
PL4: 0.00
F2 - Processing parameters
SI: 32768
SF: 101.6250000 MHz
WDW: EM
SSB: 0
GB: 0
PC: 1.40
  
```

Fig. 15: <sup>1</sup>H NMR of 1g

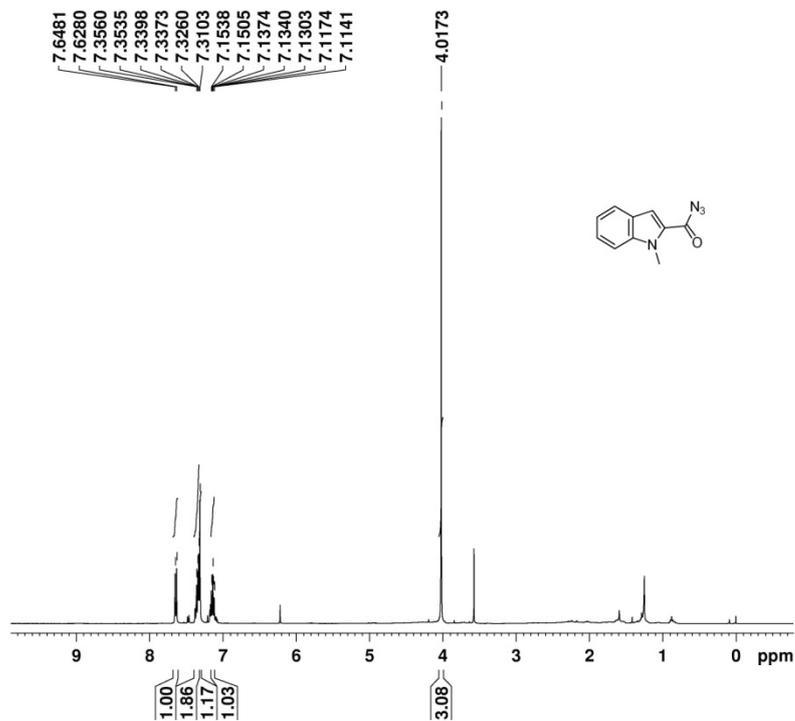


RKKB-465

```

Current Data Parameters
NAME 29-05-2014_400 FN 13C
EXPNO 440
PROCNO 1
F2 - Acquisition Parameters
DATE_ 20140529
TIME 23.14
INSTRUM spect
PROBHD 5 mm PABBO 90/
PULPROG zgpg30
TD 65536
SOLVENT CDCl3
NS 212
DS 4
SWH 24038.442 Hz
FIDRES 0.1860798 Hz
AQ 1.3521488 sec
RG 201.48
DM 220.800 usec
DE 6.50 usec
TE 300.2 K
D1 2.00000000 sec
D11 0.03000000 sec
TDO 1
  
```

Fig. 16: <sup>13</sup>C NMR of 1g

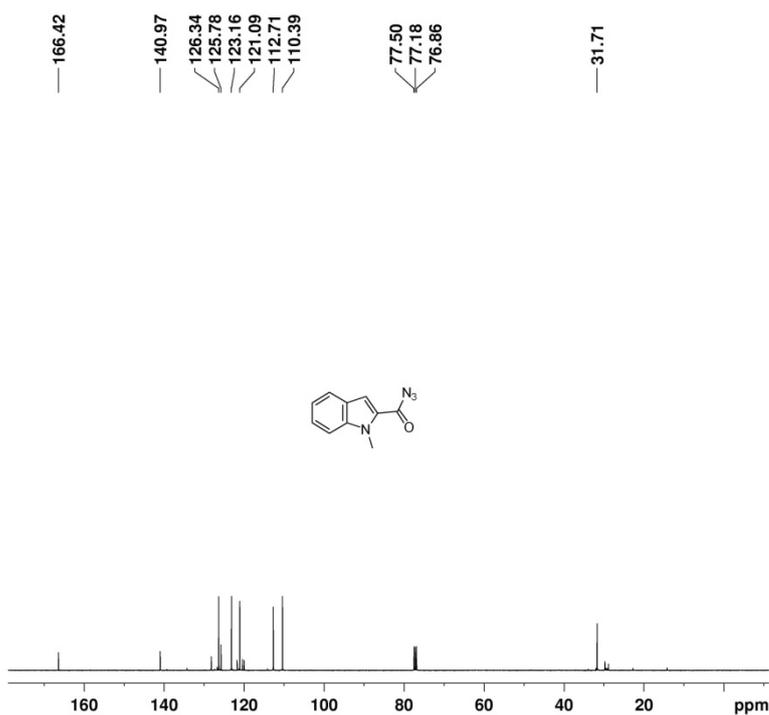


RKKB-469

```

Current Data Parameters
NAME 08-06-2014_400-1H AN
EXPNO 412
PROCNO 1
F2 - Acquisition Parameters
DATE_ 20140607
TIME 31.14
INSTRUM spect
PROBHD 5 mm PABBO 90/
PULPROG zgpg30
TD 65536
SOLVENT CDCl3
NS 212
DS 4
SWH 8012.820 Hz
FIDRES 0.122566 Hz
AQ 4.0981488 sec
RG 201.48
DM 220.800 usec
DE 6.50 usec
TE 300.2 K
D1 1.00000000 sec
TDO 1
  
```

Fig. 17: <sup>1</sup>H NMR of 11



RKBK-469

```

Current Data Parameters
NAME: 05-09-11-01 13C M
DATE_: 20140115
PROCNO: 1
F2 - Acquisition Parameters
Date_: 20140115
Time: 14.21
INSTRUM: spect
PROBHD: 5 mm PABBO BB/
PULPROG: zgpg
SOLVENT: CDCl3
NS: 8
DS: 0
SWH: 20176.100 Hz
FIDRES: 0.166338 Hz
AQ: 4.965463 sec
RG: 20.54
DM: 62.400 usec
DE: 6.50 usec
TE: 300.2 K
D1: 1.00000000 sec
TD0: 1
===== CHANNEL f1 =====
NUC1: 13C
P1: 1.20 usec
PL1: 0.00000000 W
===== CHANNEL f2 =====
NUC2: 1H
P2: 1.20 usec
PL2: 0.00000000 W
===== CHANNEL f3 =====
NUC3: 1H
P3: 1.20 usec
PL3: 0.00000000 W
F2 - Processing parameters
SI: 65536
SF: 400.1603000 MHz
WDW: EM
SSB: 0
GB: 0
PC: 1.00
  
```

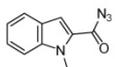
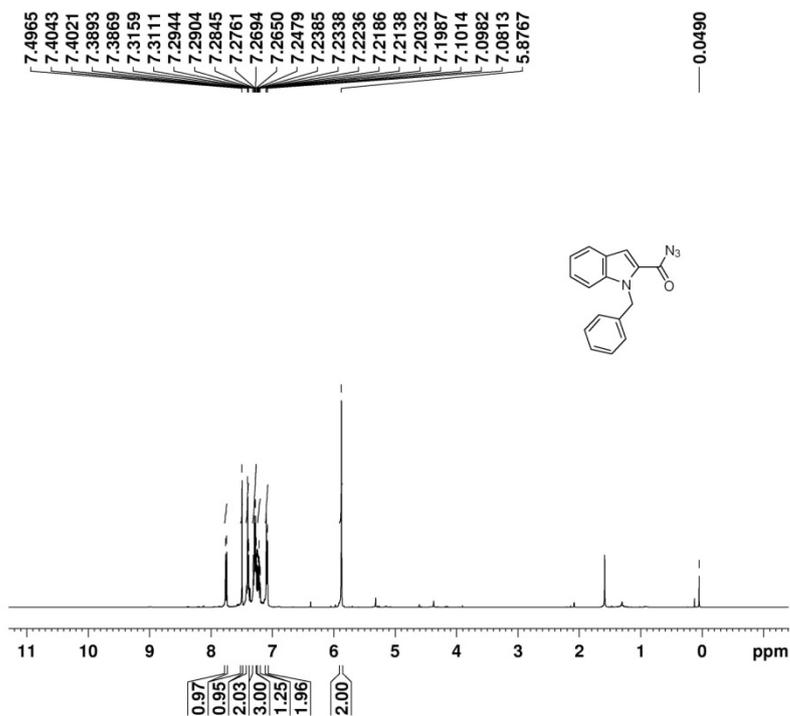


Fig. 18: <sup>13</sup>C NMR of 11



RKBK-AZZ

```

Current Data Parameters
NAME: 15-04-14 FN 18
EXPNO: 630
PROCNO: 1
F2 - Acquisition Parameters
Date_: 20140415
Time: 14.21
INSTRUM: spect
PROBHD: 5 mm PABBO BB/
PULPROG: zgpg
SOLVENT: CDCl3
NS: 8
DS: 0
SWH: 8012.820 Hz
FIDRES: 0.122266 Hz
AQ: 4.089463 sec
RG: 80.54
DM: 62.400 usec
DE: 6.50 usec
TE: 300.2 K
D1: 1.00000000 sec
TD0: 1
===== CHANNEL f1 =====
SFO1: 400.1603000 MHz
NUC1: 1H
P1: 12.35 usec
PL1: 14.00000000 W
F2 - Processing parameters
SI: 65536
SF: 400.1603000 MHz
WDW: EM
SSB: 0
GB: 0
PC: 1.00
  
```

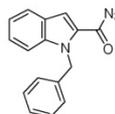
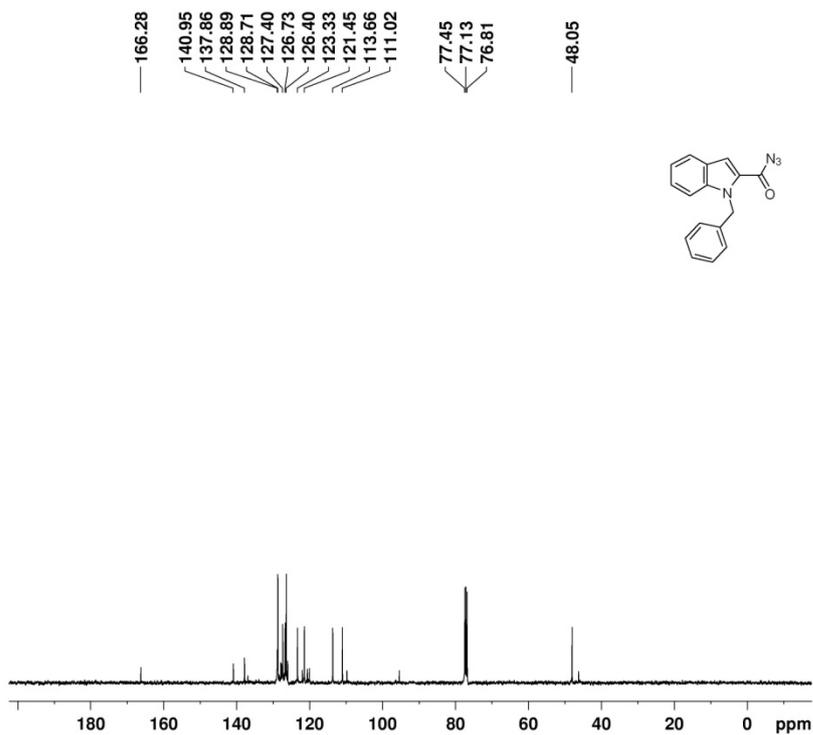


Fig. 19: <sup>1</sup>H NMR of 1m



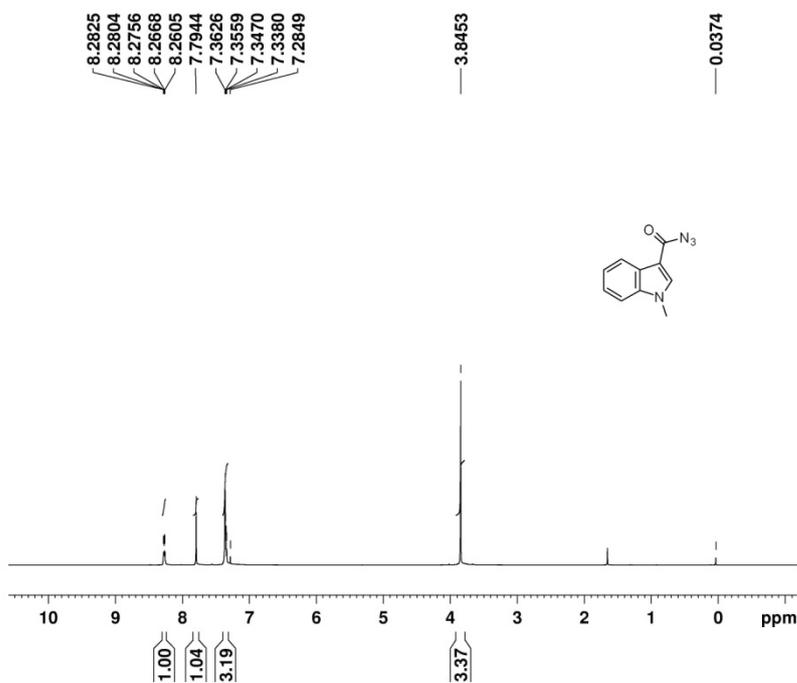
RKBK-AZ2

```

Current Data Parameters
NAME: 01-01-14_F3_14
EXPNO: 1
PROCNO: 1
PROCNAME:
PULPROG: zgpg30
TD: 65536
SOLVENT:
NS: 2
DS: 4
SWH: 16312.000 Hz
FIDRES: 0.122259 Hz
AQ: 4.098461 sec
RG: 102.41
SQ: 0.7500000 sec
TE: 300.2 K
DQ: 1.0000000 sec
TD0:
----- CHANNEL f1 -----
NUC1: 13C
P1: 12.00
PC: 1.00

```

Fig. 20: <sup>13</sup>C NMR of 1m



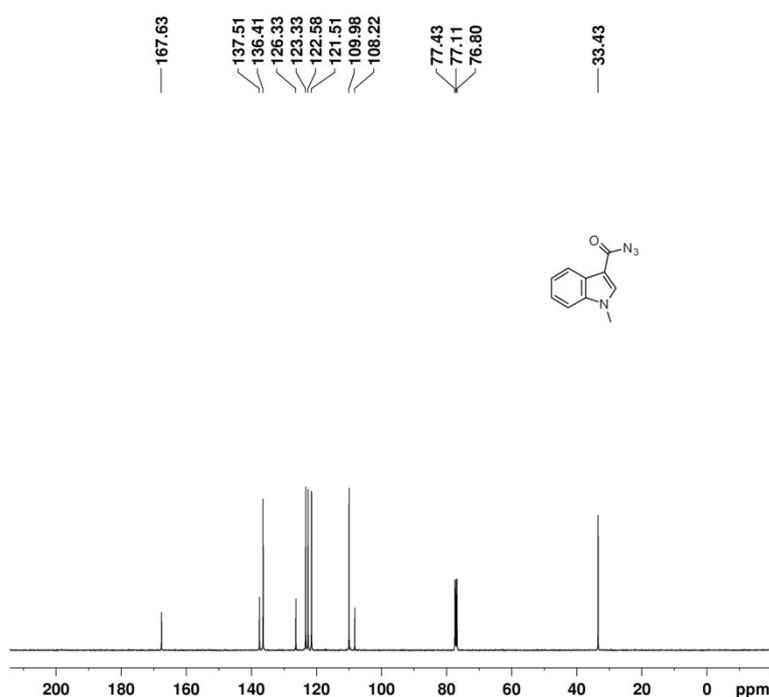
RKBK-AZ

```

Current Data Parameters
NAME: 01-01-14_F3_14
EXPNO: 1
PROCNO: 1
PROCNAME:
PULPROG: zgpg30
TD: 65536
SOLVENT:
NS: 2
DS: 4
SWH: 16312.000 Hz
FIDRES: 0.122259 Hz
AQ: 4.098461 sec
RG: 102.41
SQ: 0.7500000 sec
TE: 300.2 K
DQ: 1.0000000 sec
TD0:
----- CHANNEL f1 -----
NUC1: 1H
P1: 12.00
PC: 1.00

```

Fig. 21: <sup>1</sup>H NMR of 1n

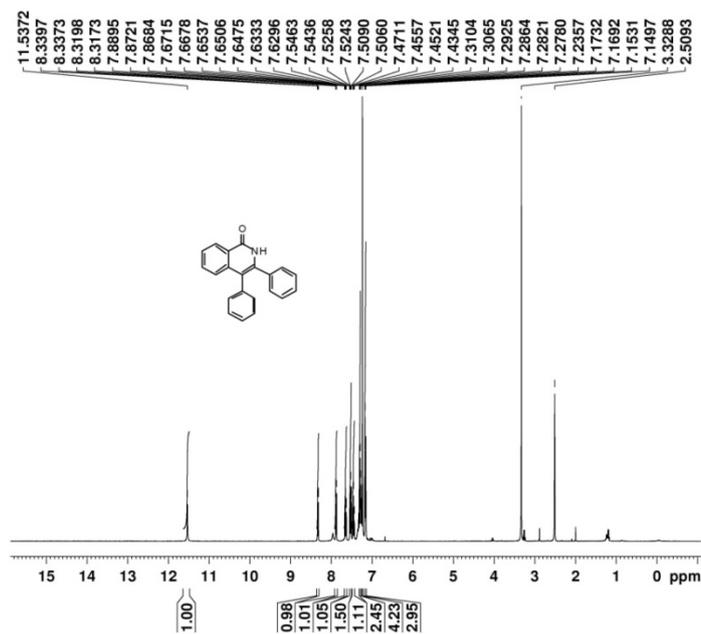


RKBC-AZ

```

Current Data Parameters
NAME: z1-03-2014-400FN
EXPNO: 2
PROCNO: 1
F2 - Acquisition Parameters
Date_: 20140321
Time: 16.55
INSTRUM: MBECL
PROBHD: 5 mm PABBO BB/
PULPROG: zgpg30
TD: 65536
SOLVENT: DMSO
NS: 0
DS: 0
SWH: 9615.385 Hz
FIDRES: 0.146719 Hz
AQ: 3.4378720 sec
RG: 134.20
EM: 32.000 usec
DE: 6.50 usec
TE: 300.0 K
D1: 1.00000000 sec
TD0: 1
----- CHANNEL f1 -----
SFO1: 400.1428712 MHz
NUC1: 13
P1: 12.65 usec
PLM1: 13.10000038 W
F2 - Processing parameters
SI: 65536
SF: 400.1400000 MHz
MCW: EM
SSB: 0
LB: 0.30 Hz
GB: 0
PC: 1.00
  
```

Fig. 22: <sup>13</sup>C NMR of 1n

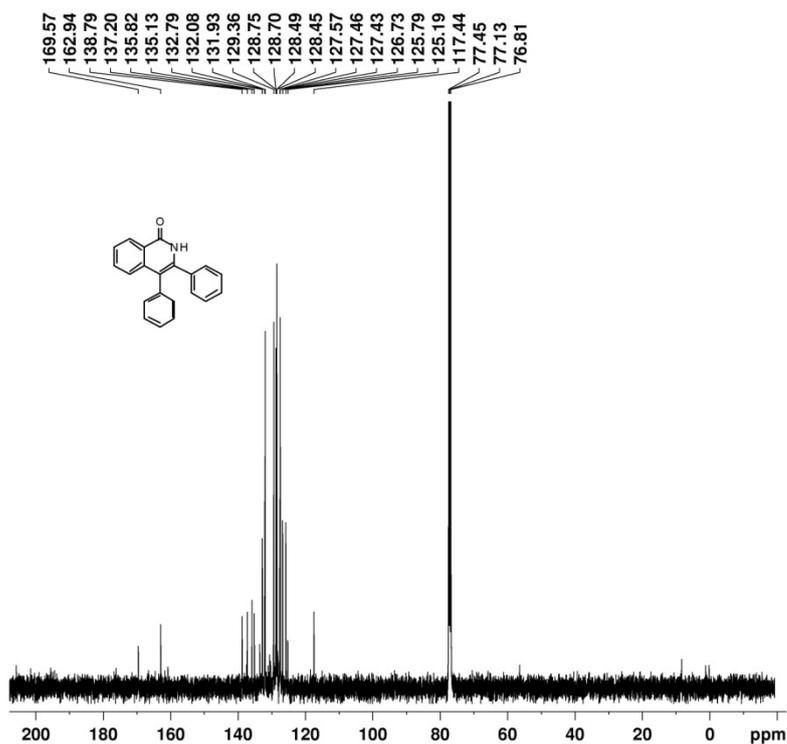


RKBC-C2

```

Current Data Parameters
NAME: z1-03-2014-400FN
EXPNO: 2
PROCNO: 1
F2 - Acquisition Parameters
Date_: 20140321
Time: 16.55
INSTRUM: MBECL
PROBHD: 5 mm PABBO BB/
PULPROG: zgpg30
TD: 65536
SOLVENT: DMSO
NS: 0
DS: 0
SWH: 9615.385 Hz
FIDRES: 0.146719 Hz
AQ: 3.4378720 sec
RG: 134.20
EM: 32.000 usec
DE: 6.50 usec
TE: 300.0 K
D1: 1.00000000 sec
TD0: 1
----- CHANNEL f1 -----
SFO1: 400.1428712 MHz
NUC1: 13
P1: 12.65 usec
PLM1: 13.10000038 W
F2 - Processing parameters
SI: 65536
SF: 400.1400000 MHz
MCW: EM
SSB: 0
LB: 0.30 Hz
GB: 0
PC: 1.00
  
```

Fig. 23: <sup>1</sup>H NMR of 4aa

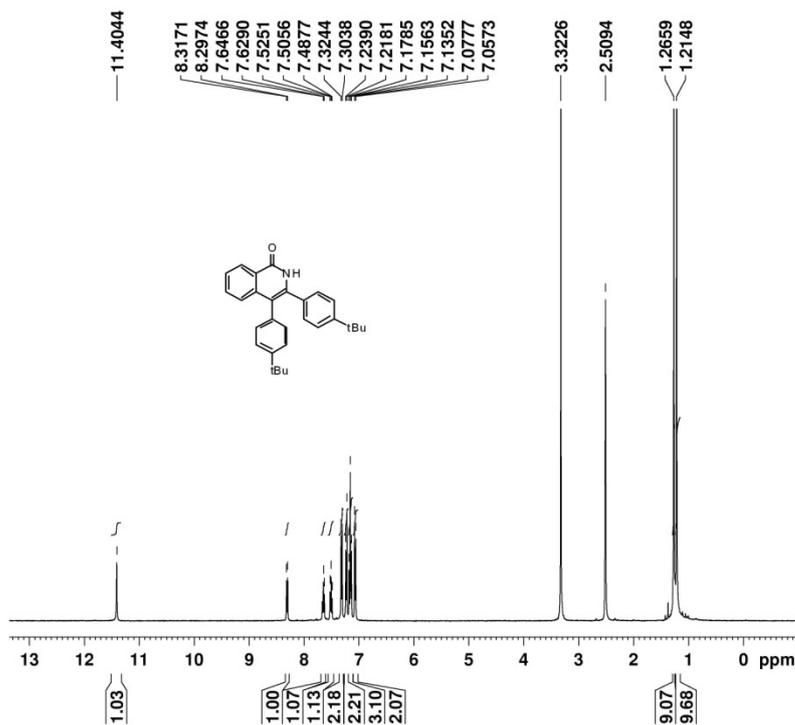


RKBK-C2

```

Current Data Parameters
NAME: 21-03-2014-005_13C AN
EXPNO: 1
PROCNO: 1
F2 - Acquisition Parameters
DATE_: 20140321
TIME: 12.35
INSTRUM: spect
PROBHD: 5 mm PABBO BB
PULPROG: zgpg30
TD: 65536
SOLVENT: CDCl3
NS: 312
DS: 4
DE: 24038.461 Hz
FIDRES: 0.288728 Hz
AQ: 1.1632488 sec
RG: 251.48
AQ: 29.480 usec
DE: 0.50 usec
TE: 300.2 K
DQ: 2.0000000 sec
SFO: 100.6261200 MHz
TD0: 1
----- CHANNEL f1 -----
NUC1: 13C
P1: 12.00 usec
PL1: 0.00 dB
PC1: 100.6261200 MHz
----- CHANNEL f2 -----
SFO2: 400.1421000 MHz
NUC2: 1H
P2: 12.00 usec
PL2: 0.00 dB
PC2: 400.1421000 MHz
F2 - Processing parameters
SI: 32768
SF: 100.6261200 MHz
WDW: EM
SSB: 0
LB: 1.00 Hz
GB: 0
PC: 1.40
  
```

Fig. 24: <sup>13</sup>C NMR of 4aa

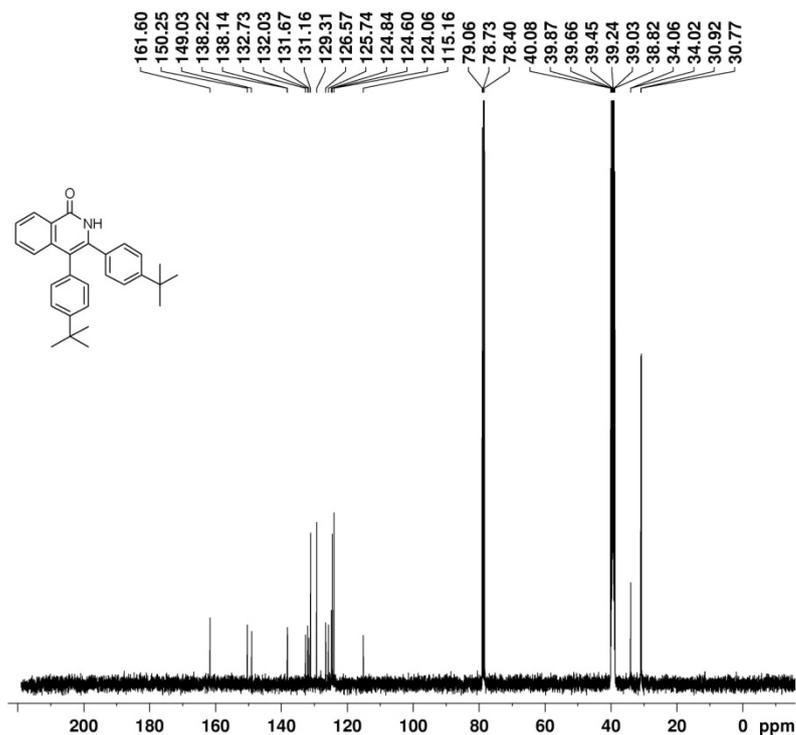


RKBK-C4

```

Current Data Parameters
NAME: 21-03-2014-005F1
EXPNO: 1
PROCNO: 1
F2 - Acquisition Parameters
DATE_: 20140321
TIME: 17.02
INSTRUM: spect
PROBHD: 5 mm PABBO BB
PULPROG: zgpg30
TD: 65536
SOLVENT: DMSO
NS: 5
DS: 5
DE: 8615.185 Hz
FIDRES: 0.146719 Hz
AQ: 1.4071920 sec
RG: 114.24
AQ: 32.000 usec
DE: 1.50 usec
TE: 300.2 K
DQ: 1.0000000 sec
SFO: 400.1421000 MHz
TD0: 1
----- CHANNEL f1 -----
NUC1: 1H
P1: 12.00 usec
PL1: 13.10000000 W
F2 - Processing parameters
SI: 32768
SF: 400.1421000 MHz
WDW: EM
SSB: 0
LB: 0.30 Hz
GB: 0
PC: 1.00
  
```

Fig. 25: <sup>1</sup>H NMR of 4ad



RKBK-C4

```

Current Data Parameters
NAME 30-09-2014-400 13ad
EXPNO 640
PROCNO 1

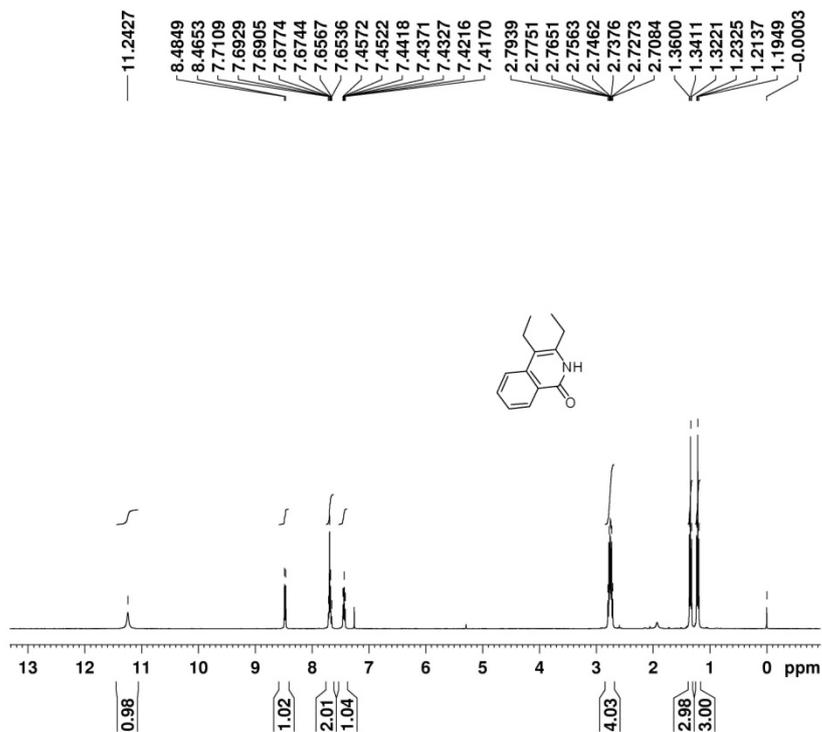
F2 - Acquisition Parameters
Date_ 20140601
Time 6.39
INSTRUM spect
PROBHD 5 mm PABBO BB
PULPROG zgpg30
TD 65536
SOLVENT DMCO
NS 2048
DS 4
SWH 24036.461 Hz
FIDRES 0.146796 Hz
AQ 1.3431488 sec
RG 301.48
DM 20.800 usec
DE 6.50 usec
TE 300.0 K
D1 2.0000000 sec
D11 0.0300000 sec
TD 1

===== CHANNEL f1 =====
NUC1 100.630493 MHz
P1 1.00 usec
PL 0.00 dB
FWD1 61.93999647 W

===== CHANNEL f2 =====
NUC2 400.142106 MHz
P2 1.00 usec
PL 0.00 dB
FWD2 247.79998574 W

F2 - Processing parameters
SI 32768
SF 100.620495 MHz
WDW EM
SSB 0
GB 0
PC 1.40
  
```

Fig. 26: <sup>13</sup>C NMR of 4ad



RKBK-434

```

Current Data Parameters
NAME 14-06-14 4ae
EXPNO 610
PROCNO 1

F2 - Acquisition Parameters
Date_ 20140615
Time 15.39
INSTRUM spect
PROBHD 5 mm PABBO BB
PULPROG zgpg30
TD 65536
SOLVENT CDCl3
NS 2048
DS 4
SWH 9613.363 Hz
FIDRES 0.146712 Hz
AQ 3.4378720 sec
RG 301.48
DM 20.800 usec
DE 6.50 usec
TE 300.0 K
D1 1.0000000 sec
TD 1

===== CHANNEL f1 =====
NUC1 400.142912 MHz
P1 1.00 usec
PL 0.00 dB
FWD1 14.00000000 W

F2 - Processing parameters
SI 32768
SF 400.142912 MHz
WDW EM
SSB 0
GB 0
PC 1.00
  
```

Fig. 27: <sup>1</sup>H NMR of 4ae

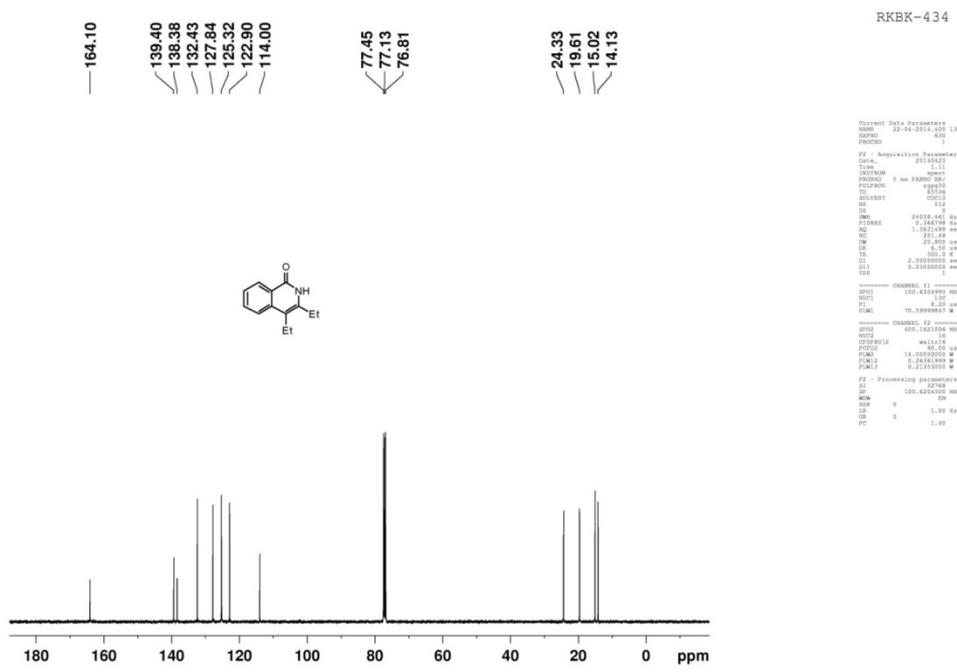


Fig. 28: <sup>13</sup>C NMR of 4ae

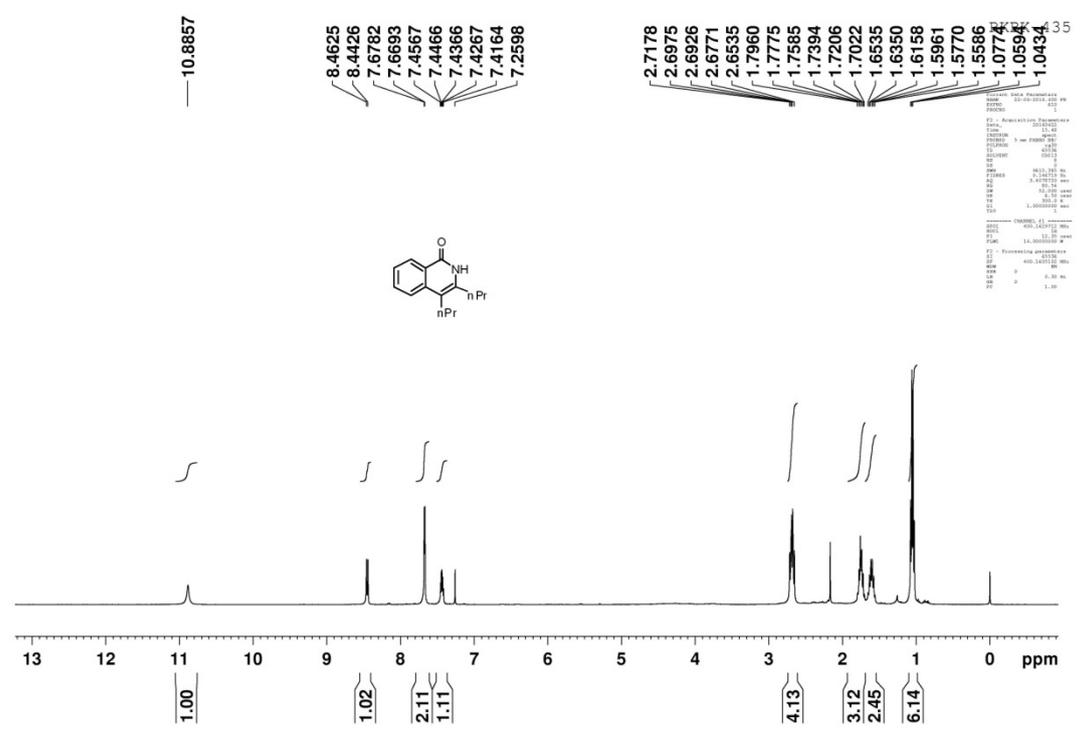
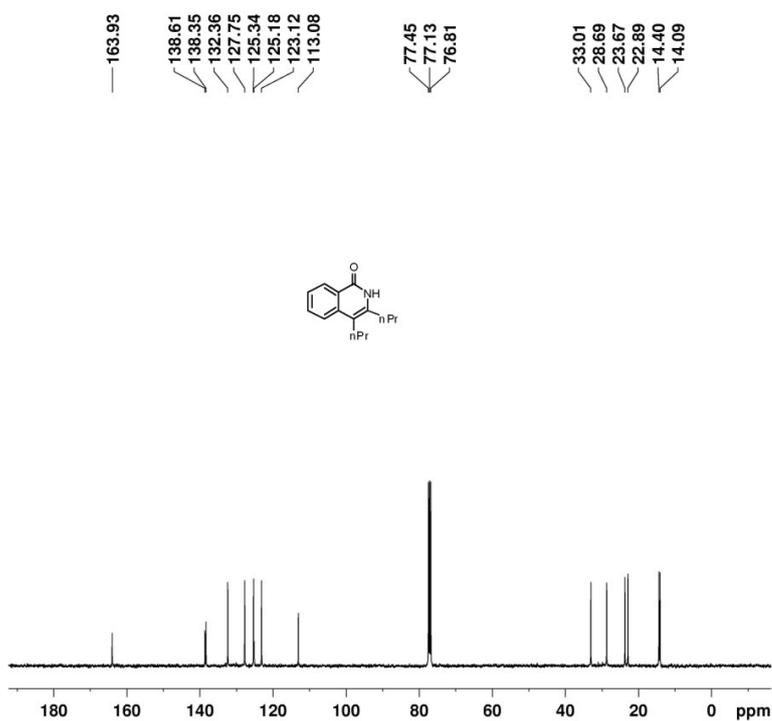


Fig. 29: <sup>1</sup>H NMR of 4af



RKKB-435

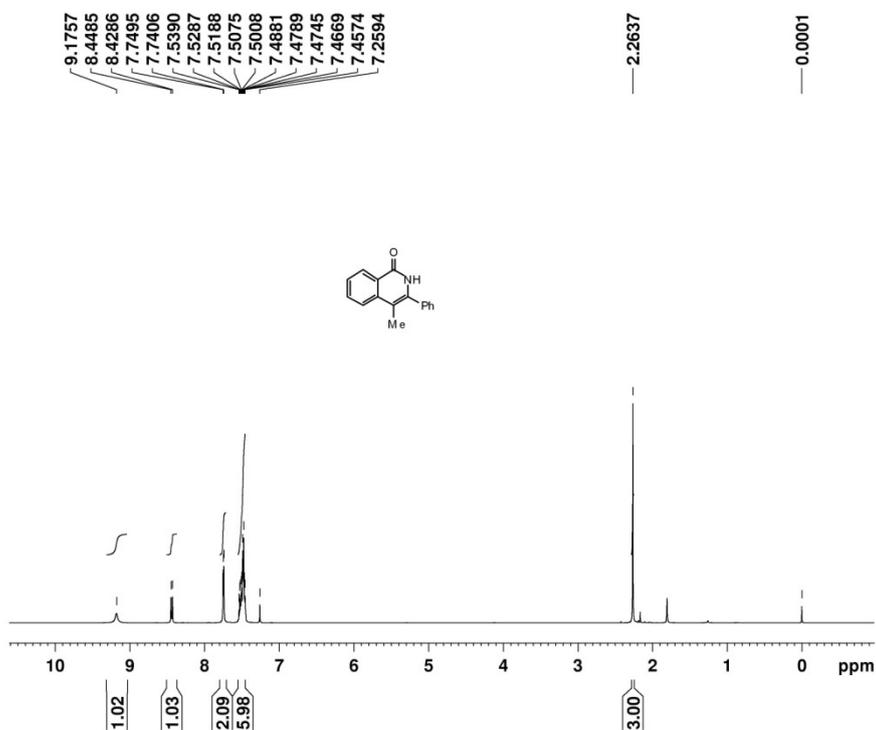
```

Current Data Parameters
NAME      24-04-2014-402 1AC FN
EXPNO    630
PROCNO   1

F2 - Acquisition Parameters
Date_    01/04/15
Time     15.10
INSTRUM  spect
PROBHD   5 mm F4002 307
PULPROG  zgpg30
TD        65536
SOLVENT  CDCl3
NS        512
DS        8
SWH       34028.441 Hz
FIDRES    0.161709 Hz
AQ        3.391149 sec
RG         256.000
WDW        EM
SS         4.10 usec
LB         0.30 Hz
GB         0
PC         1.00
===== CHANNEL f1 =====
NUC1      13C
P1        12.00 usec
PL1       0.00 dB
===== CHANNEL f2 =====
NUC2      1H
P2        12.00 usec
PL2       0.00 dB
===== CHANNEL f3 =====
NUC3      1H
P3        14.0000000 usec
PL3       0.00 dB
===== CHANNEL f4 =====
NUC4      1H
P4        14.0000000 usec
PL4       0.00 dB

F2 - Processing parameters
SI        45536
SF        400.162912 MHz
WDW        EM
SSB        0
GB         0.30 Hz
PC         1.00
  
```

Fig. 30: <sup>13</sup>C NMR of 4af



RKKB-437

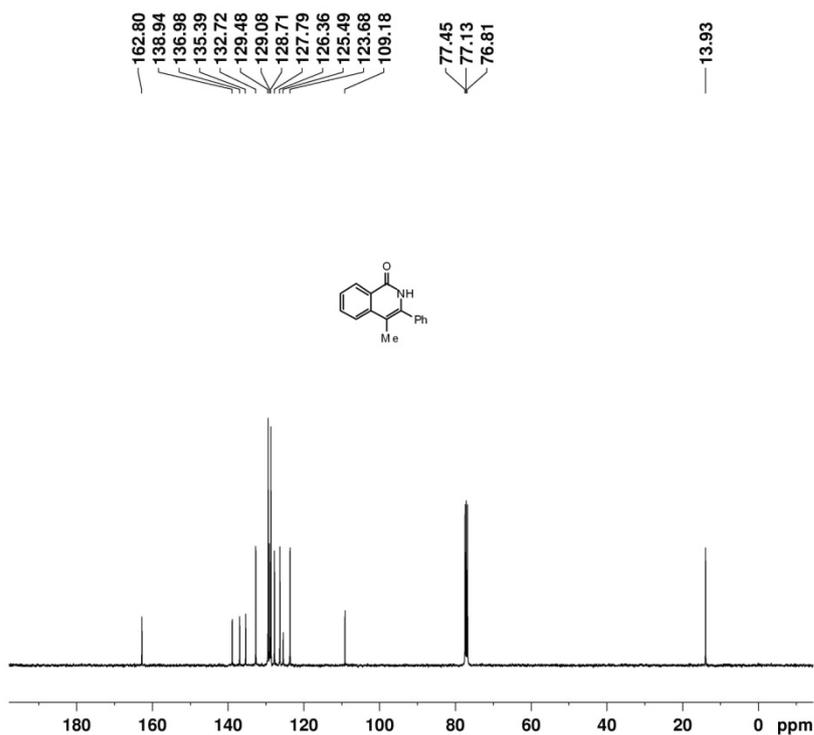
```

Current Data Parameters
NAME      22-04-2014.400 FN
EXPNO    630
PROCNO   1

F2 - Acquisition Parameters
Date_    20/10/12
Time     16.00
INSTRUM  spect
PROBHD   5 mm F4002 307
PULPROG  zgpg30
TD        65536
SOLVENT  CDCl3
NS        8
DS        8
SWH       9615.385 Hz
FIDRES    0.148719 Hz
AQ        3.4078720 sec
RG         100.61
WDW        EM
SS         52.000 usec
LB         0.30 Hz
GB         0
PC         1.00000000 sec
TSD       1

===== CHANNEL f1 =====
SF01     400.162912 MHz
NUC1      1H
P1        12.35 usec
PL1       0.00 dB
===== CHANNEL f2 =====
SF       400.1605103 MHz
WDW        EM
SSB        0
GB         0.30 Hz
PC         1.00
  
```

Fig. 31: <sup>1</sup>H NMR of 4ag



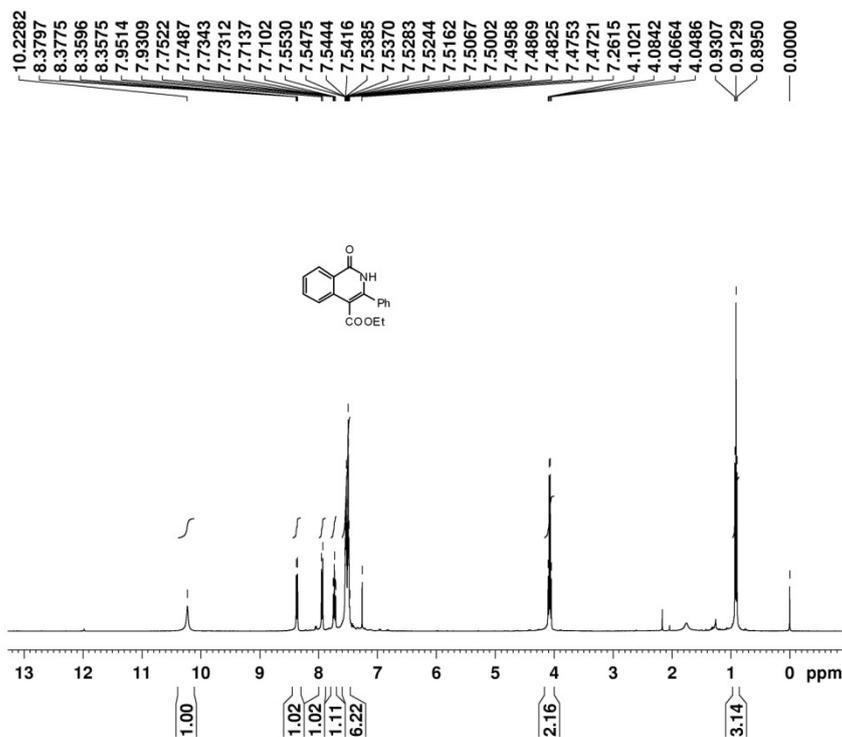
RKBK-437

```

=====
NAME          4ag
EXPNO         1
PROCNO        1
PROCNAME      4ag
F2 - Acquisition Parameters
Date_         20140424
Time          12.11
INSTRUM       spect
PROBHD        5 mm BBO-1H
PULPROG       zgpg30
SOLVENT       CDCl3
NUC1          13C
NUC2          1H
AQ            0.20000000 sec
RG            655.00000000
AQ2           0.10000000 sec
RG2           327.50000000
AQ3           0.05000000 sec
RG3           163.75000000
=====
NAME          4ag
EXPNO         1
PROCNO        1
PROCNAME      4ag
F2 - Processing parameters
SI          327.50000000
SF          101.62500000 MHz
WDW          EM
SSB          0
LB           0.30000000 Hz
GB           0
PC           1.00000000 sec
SFO          101.62500000 MHz
WDW          EM
SSB          0
LB           0.30000000 Hz
GB           0
PC           1.00000000 sec
=====

```

Fig. 32: <sup>13</sup>C NMR of 4ag



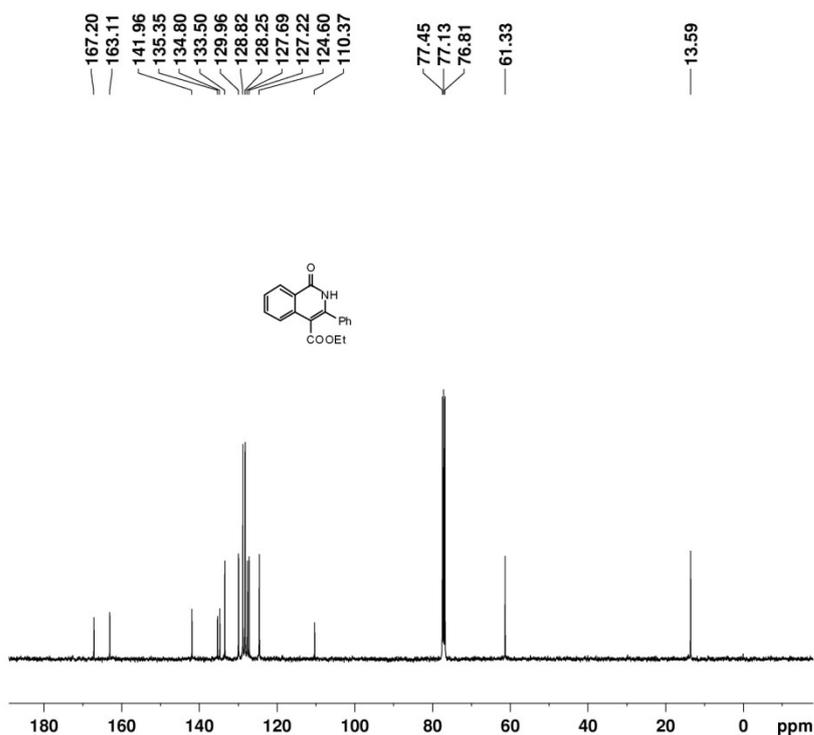
RKBK-436

```

=====
NAME          4ah
EXPNO         1
PROCNO        1
PROCNAME      4ah
F2 - Acquisition Parameters
Date_         20140424
Time          12.11
INSTRUM       spect
PROBHD        5 mm BBO-1H
PULPROG       zgpg30
SOLVENT       CDCl3
NUC1          1H
NUC2          13C
AQ            0.20000000 sec
RG            655.00000000
AQ2           0.10000000 sec
RG2           327.50000000
AQ3           0.05000000 sec
RG3           163.75000000
=====
NAME          4ah
EXPNO         1
PROCNO        1
PROCNAME      4ah
F2 - Processing parameters
SI          327.50000000
SF          101.62500000 MHz
WDW          EM
SSB          0
LB           0.30000000 Hz
GB           0
PC           1.00000000 sec
SFO          101.62500000 MHz
WDW          EM
SSB          0
LB           0.30000000 Hz
GB           0
PC           1.00000000 sec
=====

```

Fig. 33: <sup>1</sup>H NMR of 4ah

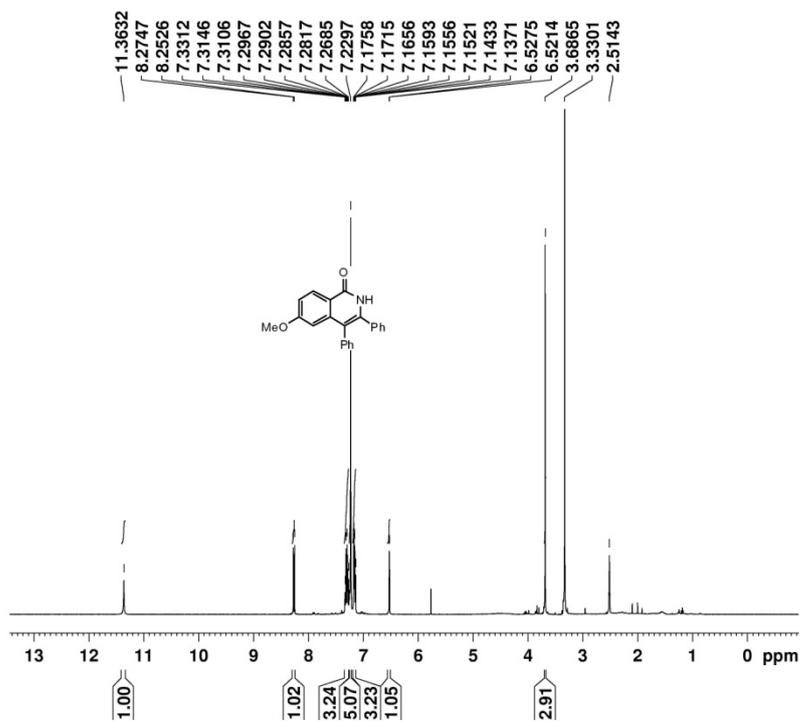


RKBK-436

```

NAME: 4ah
EXPNO: 1
PROCNO: 1
PROCPS: 1
SOLVENT: CDCl3
AQ: 0.12999999
RG: 327.50000000
RGY: 327.50000000
AQRES: 0.00000000
RGRES: 327.50000000
RGYRES: 327.50000000
F2: Acquisition Parameters
Date_ : 2014-12-10
Time: 17.18
INSTRUM: spect
PROBHD: 5 mm PABBO-1
PULPROG: zgpg30
TD: 65536
SFO: 125.76135
AQ: 0.12999999
RG: 327.50000000
RGY: 327.50000000
F2: Processing parameters
Date_ : 2014-12-10
Time: 17.22
INSTRUM: spect
PROBHD: 5 mm PABBO-1
PULPROG: zgpg30
TD: 65536
SFO: 125.76135
AQ: 0.12999999
RG: 327.50000000
RGY: 327.50000000
  
```

Fig. 34: <sup>13</sup>C NMR of 4ah

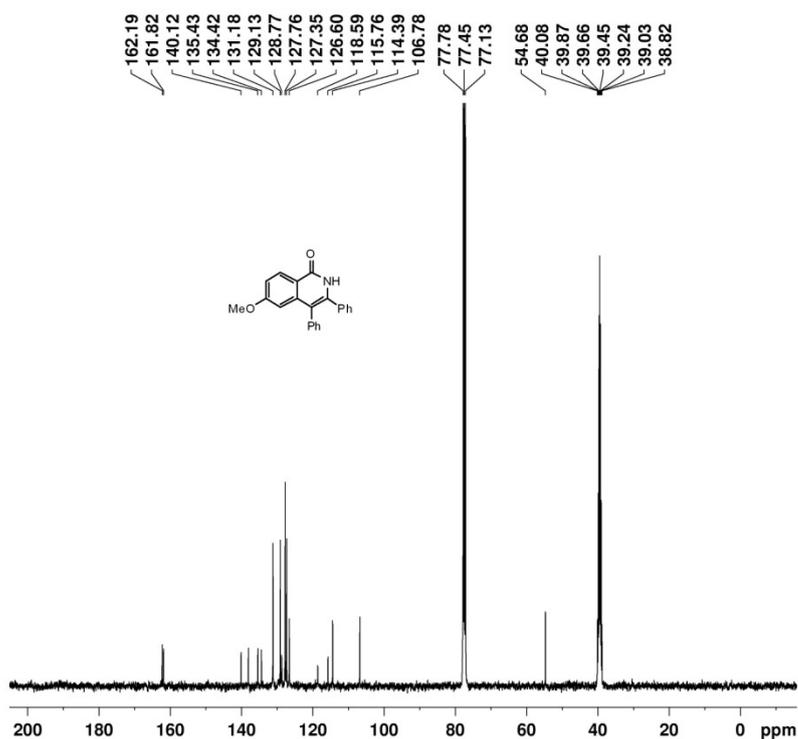


RKBK-438

```

NAME: 4ba
EXPNO: 1
PROCNO: 1
PROCPS: 1
SOLVENT: CDCl3
AQ: 0.12999999
RG: 327.50000000
RGY: 327.50000000
AQRES: 0.00000000
RGRES: 327.50000000
RGYRES: 327.50000000
F2: Acquisition Parameters
Date_ : 2014-12-10
Time: 17.18
INSTRUM: spect
PROBHD: 5 mm PABBO-1
PULPROG: zgpg30
TD: 65536
SFO: 125.76135
AQ: 0.12999999
RG: 327.50000000
RGY: 327.50000000
F2: Processing parameters
Date_ : 2014-12-10
Time: 17.22
INSTRUM: spect
PROBHD: 5 mm PABBO-1
PULPROG: zgpg30
TD: 65536
SFO: 125.76135
AQ: 0.12999999
RG: 327.50000000
RGY: 327.50000000
  
```

Fig. 35: <sup>1</sup>H NMR of 4ba



RKBK-438

```

Current Data Parameters
NAME 13-05-2014_400
EXPNO 1
PROCNO 1

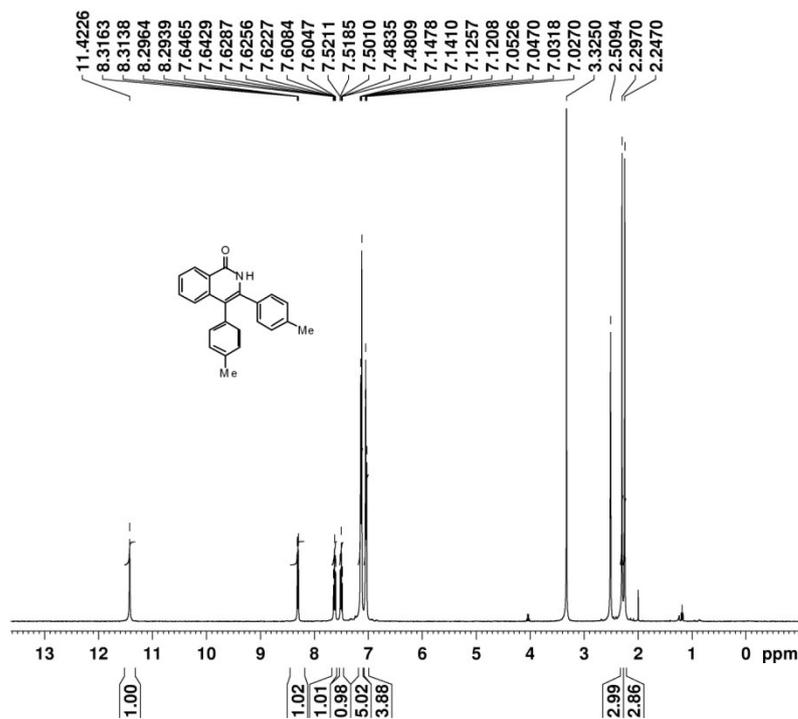
F2 - Acquisition Parameters
Date_ 20140514
Time 9.56
INSTRUM spect
PROBHD 5 mm PABBO BBP
PULPROG zgpg30
TD 65536
SOLVENT CDCl3
SI 1624
DS 2
SWE 24038.463 Hz
FIDRES 0.146798 Hz
AQ 1.163188 sec
RG 201.48
DSW 40.800 usec
DE 6.50 usec
TE 298.1 K
D1 2.0000000 sec
D11 0.0300000 sec
TDD 1

===== CHANNEL f1 =====
SFO1 100.6209147 MHz
NUC1 13C
P1 9.00 usec
PL1 0.00 dB
PLW1 61.0999947 W

===== CHANNEL f2 =====
SFO2 400.1421006 MHz
NUC2 1H
PCPD2 1H
CPCPD2 waltz16
PCPD2 90.00 usec
PLW2 13.1000038 W
PLW3 0.2470500 W
PLW3 0.2143039 W

F2 - Processing parameters
SI 32768
SF 100.6209147 MHz
WDW EM
SSB 0
GB 0
PC 1.40
  
```

Fig. 36:  $^{13}\text{C}$  NMR of 4ba



RKBK-C3

```

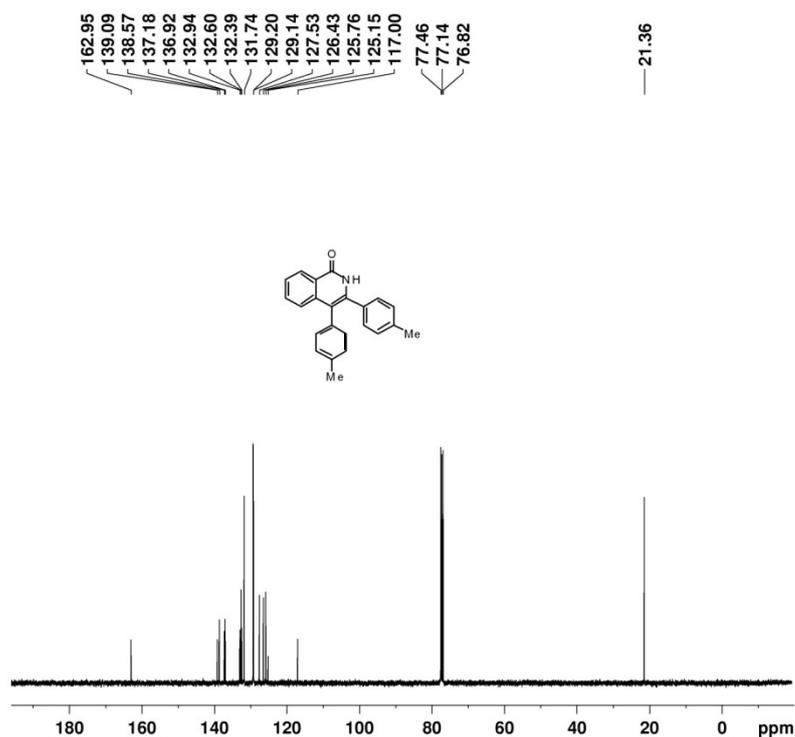
Current Data Parameters
NAME 21-03-2014_400MHz
EXPNO 1
PROCNO 1

F2 - Acquisition Parameters
Date_ 20140321
Time 16.29
INSTRUM spect
PROBHD 5 mm PABBO BBP
PULPROG zgpg30
TD 65536
SOLVENT DMF-d7
SI 1624
DS 0
SWE 9415.285 Hz
FIDRES 0.146718 Hz
AQ 3.407872 sec
RG 201.48
DSW 40.800 usec
DE 6.50 usec
TE 300.2 K
D1 1.0000000 sec
TDD 1

===== CHANNEL f1 =====
SFO1 400.1429712 MHz
NUC1 1H
P1 12.83 usec
PL1 0.00 dB
PLW1 13.1000038 W

F2 - Processing parameters
SI 32768
SF 400.1429712 MHz
WDW EM
SSB 0
GB 0
PC 1.00
  
```

Fig. 37:  $^1\text{H}$  NMR of 4ab

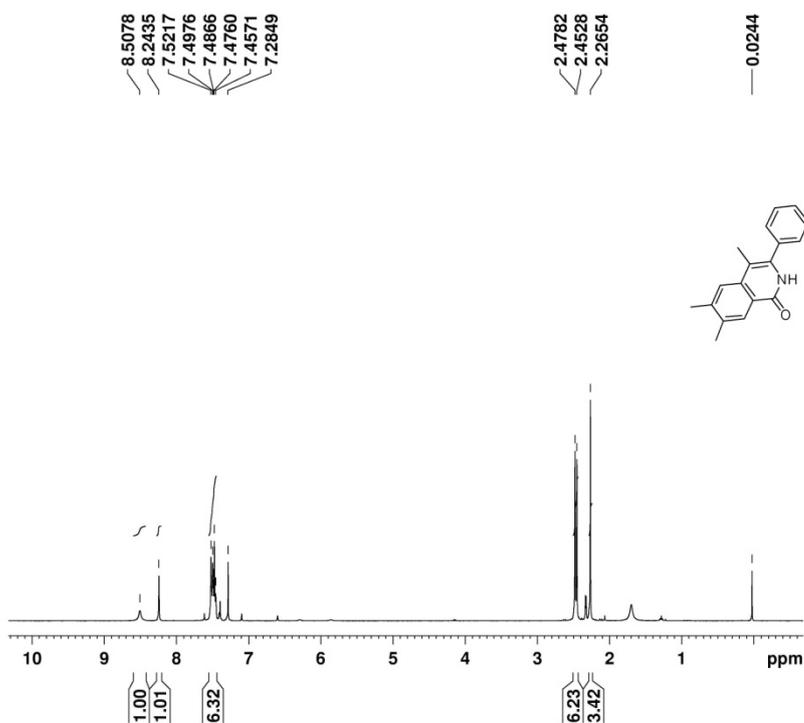


RK-C3

```

Current Data Parameters
NAME 23-04-2014.400 13C NMR
EXPNO 1
PROCNO 1
F2 - Acquisition Parameters
Date_ 20140423
Time 18.14
INSTRUM spect
PROBHD 5 mm PABBO 501
PULPROG zgpg30
SOLVENT CDCl3
NS 219
DS 4
SWH 8028.80 Hz
FIDRES 0.264780 Hz
AQ 1.932746 sec
RG 512
AQ 1.932746 sec
DE 6.50 usec
TE 300.2 K
DQ 0.0000000 sec
DEL 0.0300000 sec
SOL 1
===== CHANNEL f1 =====
NUC1 13C
P1 130
NUC2 13C
P2 130
===== CHANNEL f2 =====
SFO2 400.1421028 MHz
NUC2 13C
P2 130
===== CHANNEL f3 =====
SFO3 100.6204228 MHz
===== Processing parameters =====
SI 32768
SF 400.1405000 MHz
WDW EM
SSB 0
GB 0
PC 1.40
  
```

Fig. 38: <sup>13</sup>C NMR of 4ab

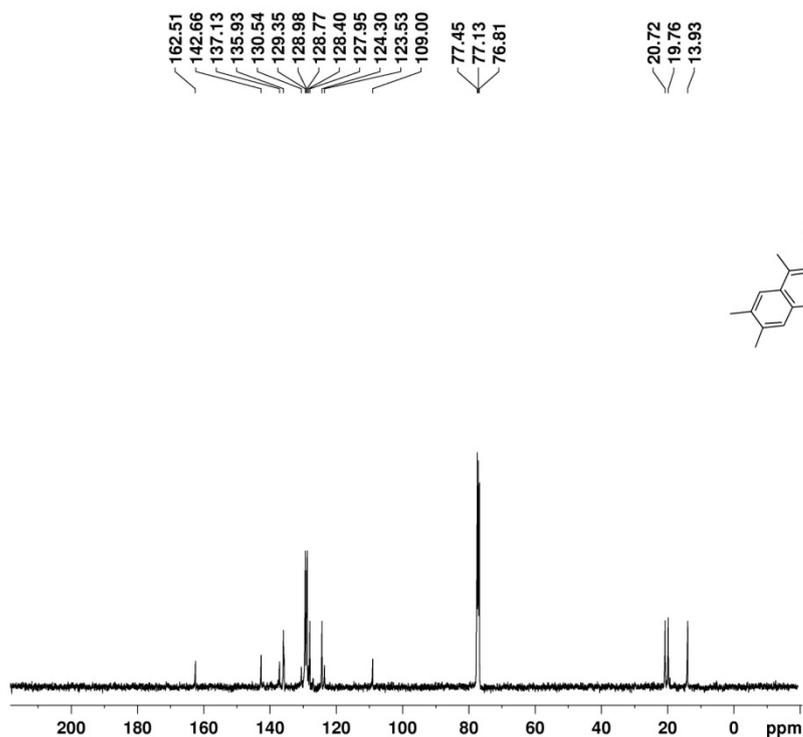


RKBK-444

```

Current Data Parameters
NAME 23-04-2014.400 400 MHz 1H NMR
EXPNO 1
PROCNO 1
F2 - Acquisition Parameters
Date_ 20140423
Time 18.14
INSTRUM spect
PROBHD 5 mm PABBO 501
PULPROG zgpg30
SOLVENT CDCl3
NS 2
DS 4
SWH 8012.600 Hz
FIDRES 0.122288 Hz
AQ 4.288465 sec
RG 512
AQ 4.288465 sec
DE 6.50 usec
TE 300.2 K
DQ 0.0000000 sec
DEL 1.0000000 sec
SOL 1
===== CHANNEL f1 =====
SFO1 400.1421028 MHz
NUC1 1H
P1 12.15 usec
P2 14.0000000 M
===== Processing parameters =====
SI 32768
SF 400.1405000 MHz
WDW EM
SSB 0
GB 0
PC 1.00
  
```

Fig. 39: <sup>1</sup>H NMR of 4bg

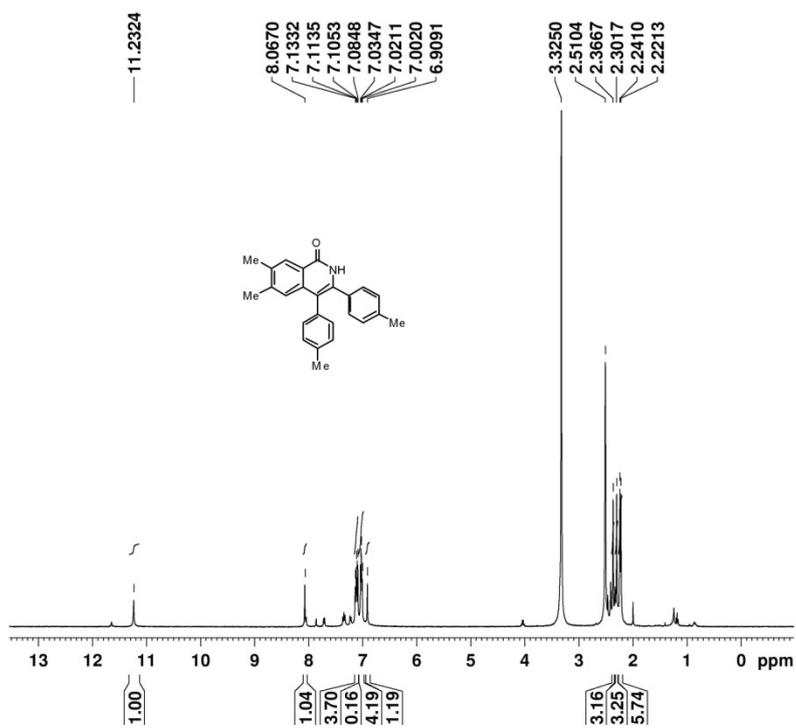


RKKB-444

```

===== CHANNEL F1 =====
Date_      20140324
Time       15.51
INSTRUM    spect
PROBHD     5 mm PABBO BB/
PULPROG    zgpg30
TD         65536
SOLVENT    DMSO
NS         8
DS         2
SWH         8012.820 Hz
FIDRES     0.122246 Hz
AQ         4.0894445 sec
RG         120.43
DM         62.400 usec
DE         6.40 usec
TE         300.0 K
D1         1.00000000 sec
TD0        1
===== CHANNEL F1 =====
SFO1       400.142912 MHz
P1         18 usec
P1M1       13.1000038 W
F2 - Processing parameters
SI         65536
SF         400.160000 MHz
WDW        EM
SSB        0
LB         0.30 Hz
GB         0
PC         1.00
  
```

Fig. 40: <sup>13</sup>C NMR of 4bg

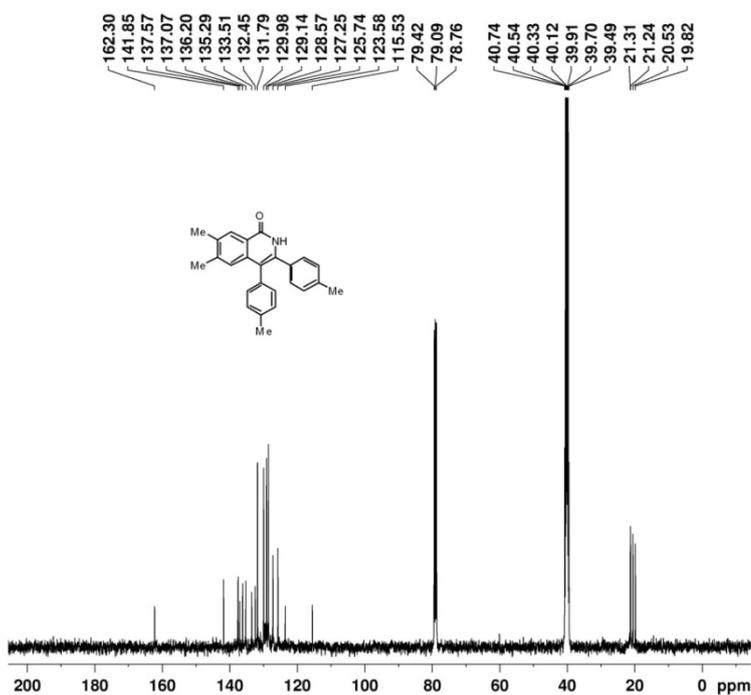


RKKB-C5

```

Current Data Parameters
NAME       21-03-2014_400 AN
EXPNO     1
PROCNO    1
F2 - Acquisition Parameters
Date_      20140324
Time       15.51
INSTRUM    spect
PROBHD     5 mm PABBO BB/
PULPROG    zgpg30
TD         65536
SOLVENT    DMSO
NS         8
DS         2
SWH         8012.820 Hz
FIDRES     0.122246 Hz
AQ         4.0894445 sec
RG         120.43
DM         62.400 usec
DE         6.40 usec
TE         300.0 K
D1         1.00000000 sec
TD0        1
===== CHANNEL F1 =====
SFO1       400.142912 MHz
P1         18 usec
P1M1       13.1000038 W
F2 - Processing parameters
SI         65536
SF         400.160000 MHz
WDW        EM
SSB        0
LB         0.30 Hz
GB         0
PC         1.00
  
```

Fig. 41: <sup>1</sup>H NMR of 4cb

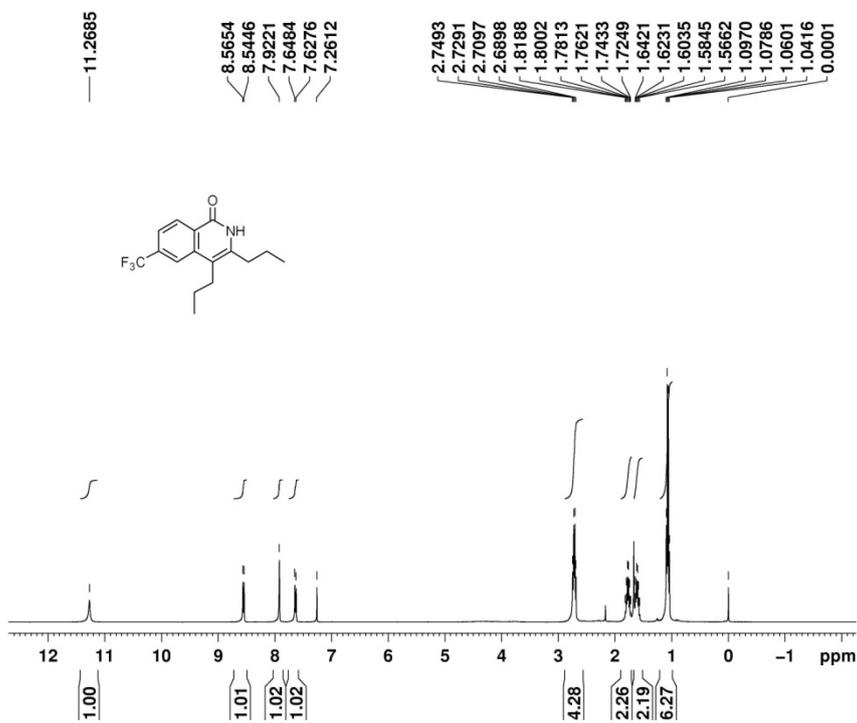


RKBK-C5

```

Current Data Parameters
NAME: 13-00-1014-000 13C
EXPNO: 1
PROCNO: 1
F2 - Acquisition Parameters
Date_ : 010101
Time: 10.40
INSTRUM: spect
PROBHD: 5 mm QNP1H/1
PULPROG: zgpg30
AQ: 0.02000000
RG: 655.00
SOLVENT: CDCl3
NS: 1024
DS: 4
SWH: 18199.620 Hz
FIDRES: 1.1811190 Hz
AQRES: 0.00010000
RG2: 655.00
SFO: 125.760000 MHz
NUC1: 13C
NUC2: 1H
PCPD2: 13.00000000 MHz
PCPD1: 0.00000000 MHz
PCPD3: 0.00000000 MHz
F2 - Processing parameters
SI: 32768
SF: 125.760000 MHz
WDW: EM
SSB: 0
LB: 3.00 Hz
GB: 0
PC: 1.40
  
```

Fig. 42: <sup>13</sup>C NMR of 4cb

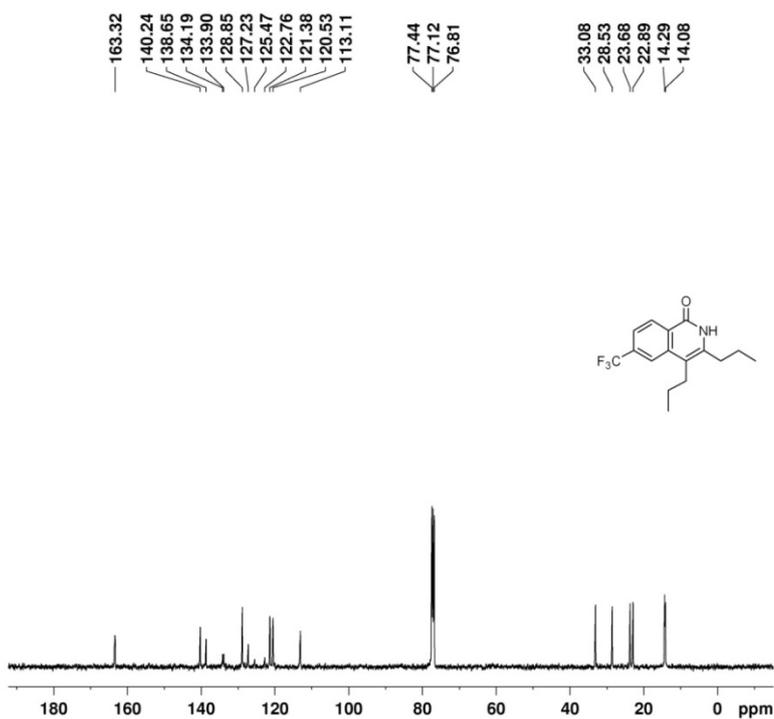


RKBK-440

```

Current Data Parameters
NAME: 13-04-1014-000 1H
EXPNO: 1
PROCNO: 1
F2 - Acquisition Parameters
Date_ : 010101
Time: 10.14
INSTRUM: spect
PROBHD: 5 mm BBOCP
PULPROG: zgpg30
AQ: 0.02000000
RG: 655.00
SOLVENT: CDCl3
NS: 1024
DS: 4
SWH: 18199.620 Hz
FIDRES: 0.1467190 Hz
AQRES: 0.00010000
RG2: 655.00
SFO: 500.136000 MHz
NUC1: 1H
NUC2: 13C
PCPD2: 13.00000000 MHz
PCPD1: 0.00000000 MHz
PCPD3: 0.00000000 MHz
F2 - Processing parameters
SI: 32768
SF: 500.136000 MHz
WDW: EM
SSB: 0
LB: 3.00 Hz
GB: 0
PC: 1.40
  
```

Fig. 43: <sup>1</sup>H NMR of 4cf

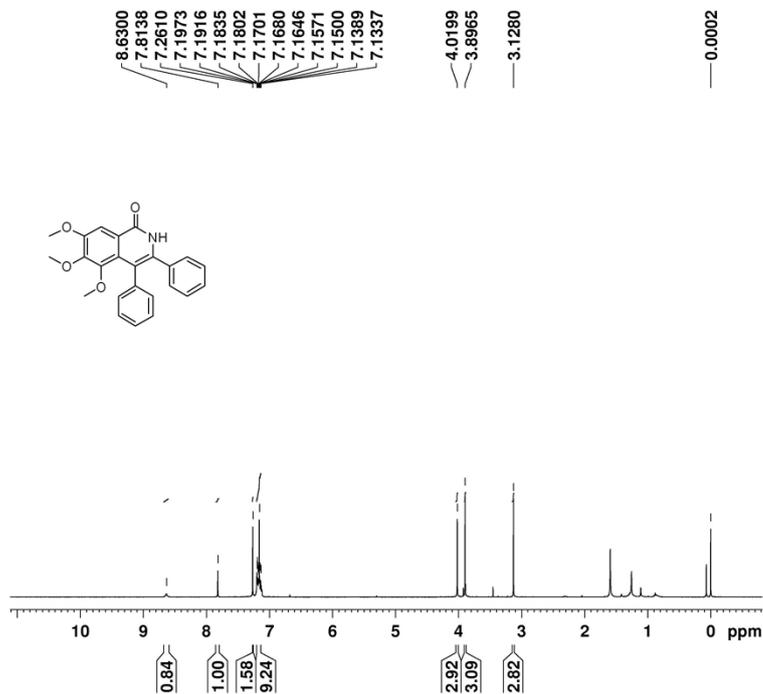


RKKB-440

```

Current Data Parameters
NAME: 27-08-2014_410
EXPNO: 1
PROCNO: 1
F2 - Acquisition Parameters
Date_ 20140827
Time 17.14
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zg30
TD 65536
SOLVENT CDCl3
NS 8
DS 0
SWH 9615.385 Hz
FIDRES 0.146719 Hz
AQ 3.4578720 sec
RG 345.29
RM 32.490 usec
DE 6.50 usec
TE 300.0 K
D1 1.00000000 sec
TD0 1
===== CHANNEL f1 =====
RF01 400.1629712 MHz
NUC1 13C
P1 12.85 usec
PIW1 13.10000038 W
F2 - Processing parameters
SI 65536
SF 400.1605995 MHz
MCW 500
SSB 0
LA 0.30 Hz
GB 0
PC 1.00
  
```

Fig. 44: <sup>13</sup>C NMR of 4cf



RKKB-432

```

Current Data Parameters
NAME: 27-08-2014_410
EXPNO: 1
PROCNO: 1
F2 - Acquisition Parameters
Date_ 20140827
Time 17.14
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zg30
TD 65536
SOLVENT CDCl3
NS 8
DS 0
SWH 9615.385 Hz
FIDRES 0.146719 Hz
AQ 3.4578720 sec
RG 345.29
RM 32.490 usec
DE 6.50 usec
TE 300.0 K
D1 1.00000000 sec
TD0 1
===== CHANNEL f1 =====
RF01 400.1629712 MHz
NUC1 13C
P1 12.85 usec
PIW1 13.10000038 W
F2 - Processing parameters
SI 65536
SF 400.1605995 MHz
MCW 500
SSB 0
LA 0.30 Hz
GB 0
PC 1.00
  
```

Fig. 45: <sup>1</sup>H NMR of 4da

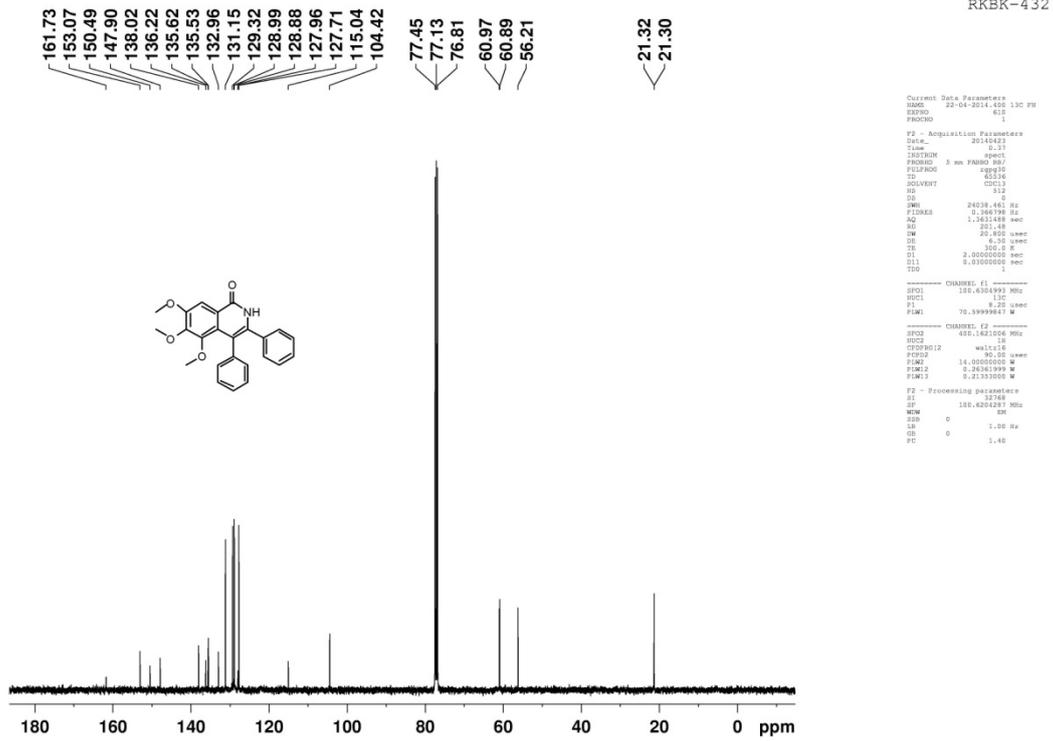
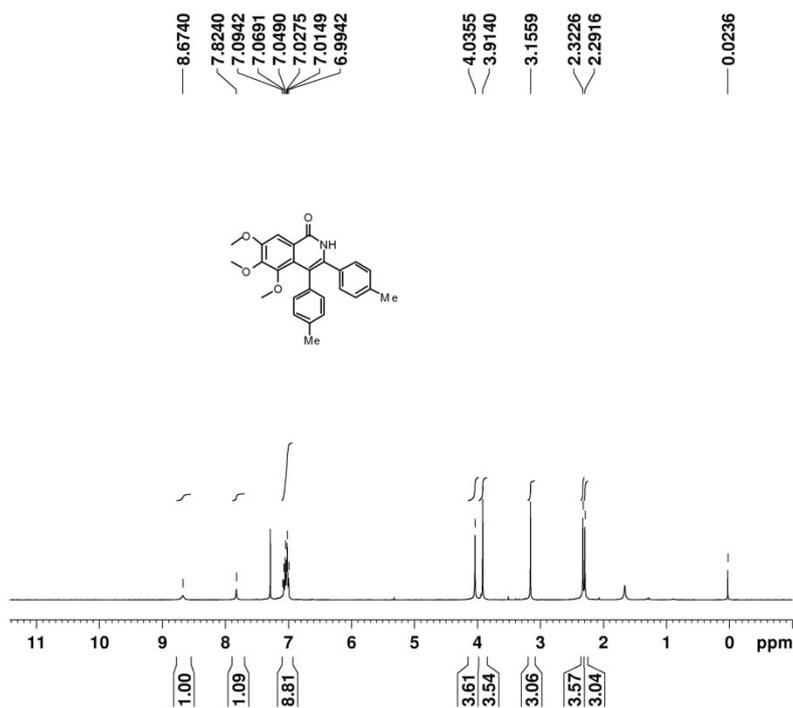


Fig. 46: <sup>13</sup>C NMR of 4da

RKBE-433

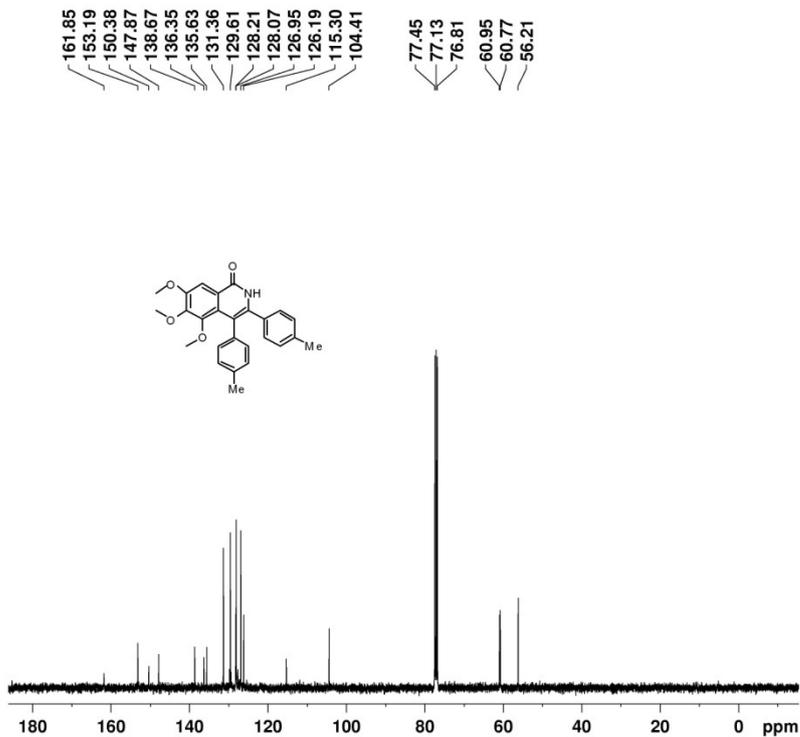


```

Current Data Parameters
NAME: 15-04-04_02_13C
EXPNO: 2
PROCNO: 1
F2 - Acquisition Parameters
Date_ : 201411
Time: 16.14
INSTRUM: spect
PROBHD: 5 mm QNP1H2
PULPROG: zgpg30
DC: 6334
SOLVENT: CDCl3
DE: 8
DWE: 802.800 Hz
FIDRES: 0.123764 Hz
AQ: 4.039160 sec
RG: 125.00
AQW: 62.400 usec
DE: 63.30 usec
SI: 819.0 Hz
D1: 1.8000000 sec
TD: 65536
===== CHANNEL f1 =====
NUC1: 13C
P1: 12.00 usec
PL1: 0.0000000 W
F2 - Processing parameters
SI: 65536
SF: 400.1400000 MHz
WDW: EM
SSB: 0
LB: 0.30 Hz
GB: 0
PC: 1.00
  
```

Fig. 47: <sup>1</sup>H NMR of 4db

RKBE-433



```

Current Data Parameters
NAME: 15-04-04_02_13C
EXPNO: 2
PROCNO: 1
F2 - Acquisition Parameters
Date_ : 201411
Time: 14.23
INSTRUM: spect
PROBHD: 5 mm QNP1H2
PULPROG: zgpg30
DC: 6334
SOLVENT: CDCl3
DE: 8
DWE: 802.800 Hz
FIDRES: 0.123764 Hz
AQ: 4.039160 sec
RG: 125.00
AQW: 62.400 usec
DE: 63.30 usec
SI: 819.0 Hz
D1: 1.8000000 sec
TD: 65536
===== CHANNEL f1 =====
NUC1: 13C
P1: 12.00 usec
PL1: 0.0000000 W
F2 - Processing parameters
SI: 65536
SF: 100.626126 MHz
WDW: EM
SSB: 0
LB: 0.30 Hz
GB: 0
PC: 1.00
  
```

Fig. 48: <sup>13</sup>C NMR of 4db

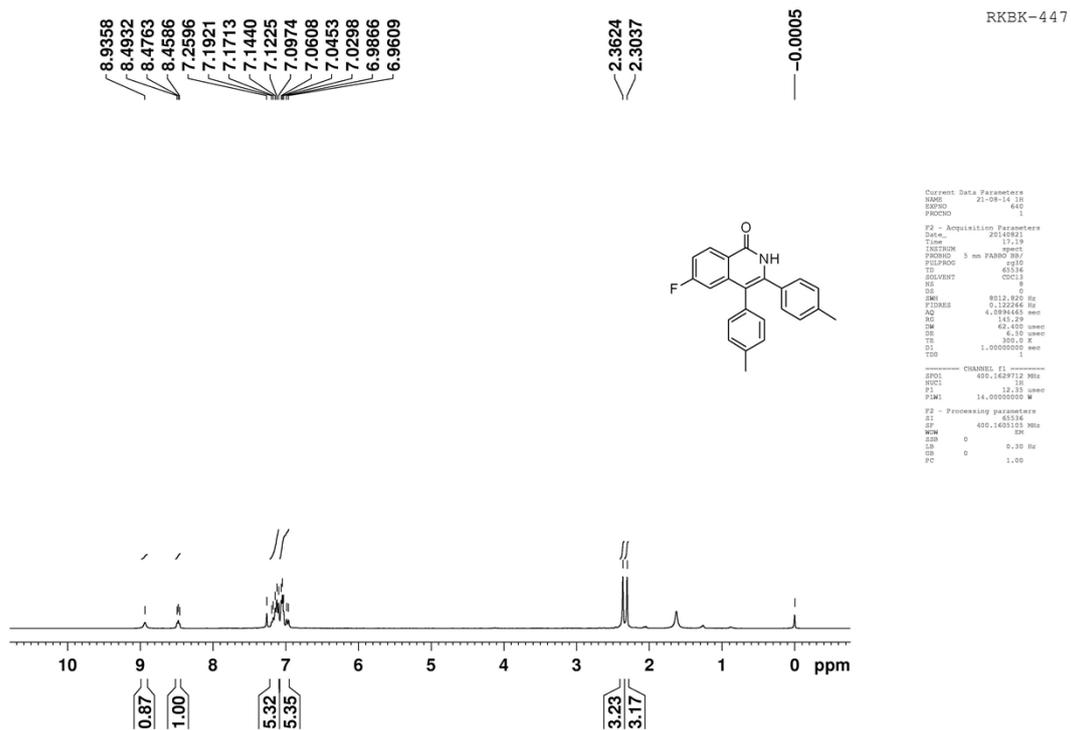


Fig. 49: <sup>1</sup>H NMR of 4eb

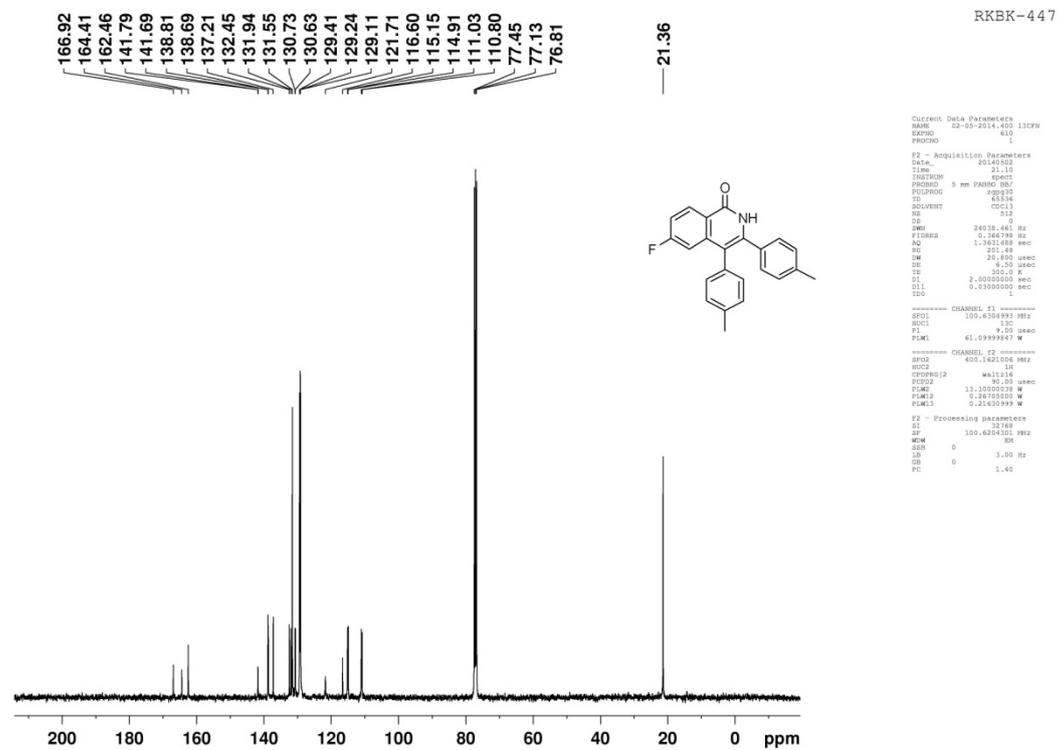
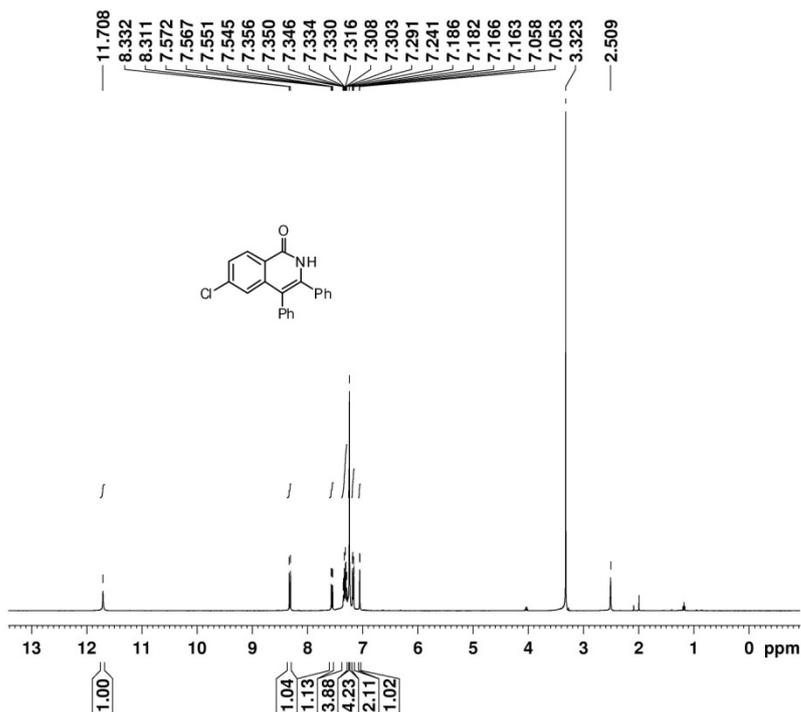


Fig. 50: <sup>13</sup>C NMR of 4eb

RKBK-441



```

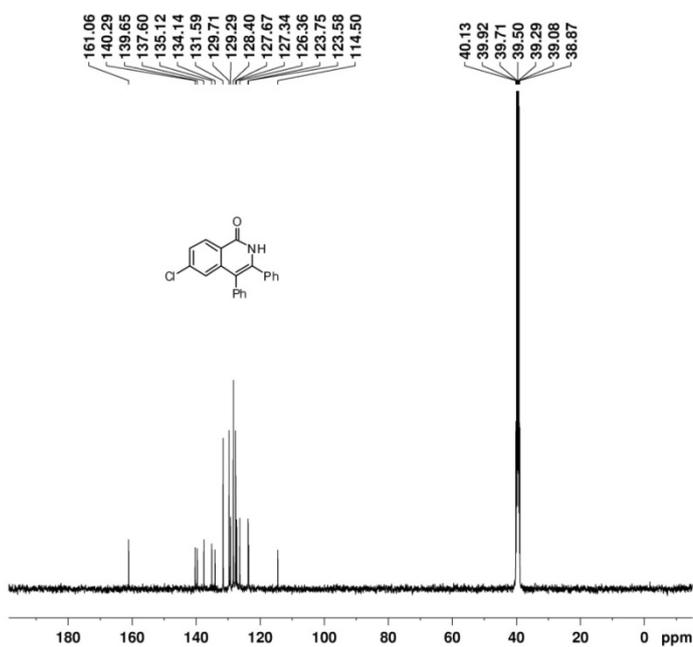
Current Data Parameters
NAME 22-04-2514.400 AN
EXPNO 610
PROCNO 1

F2 - Acquisition Parameters
DATE_ 2016022
TIME 25.42
INSTRUM spect
PROBHD 5 mm PABBO 50/
PULPROG zg30
ID 65516
SOLVENT DMF
NS 2
DS 2
SWH 8012.600 Hz
FIDRES 0.122266 Hz
AQ 4.097465 sec
RG 14.26
AQ 57.400 usec
DE 1.00 usec
TE 300.2 K
SI 1.0000000 sec
TOP

===== CHANNEL f1 =====
NUC1 400.1429712 MHz
P1 12.10 usec
PL1 0.0000000 dB
PLM1 14.0000000 dB

F2 - Processing parameters
SI 65516
SF 400.1430000 MHz
WDW EM
SSB 0
GB 0
PC 1.00
  
```

Fig. 51: <sup>1</sup>H NMR of 4fa



RKBK-441

```

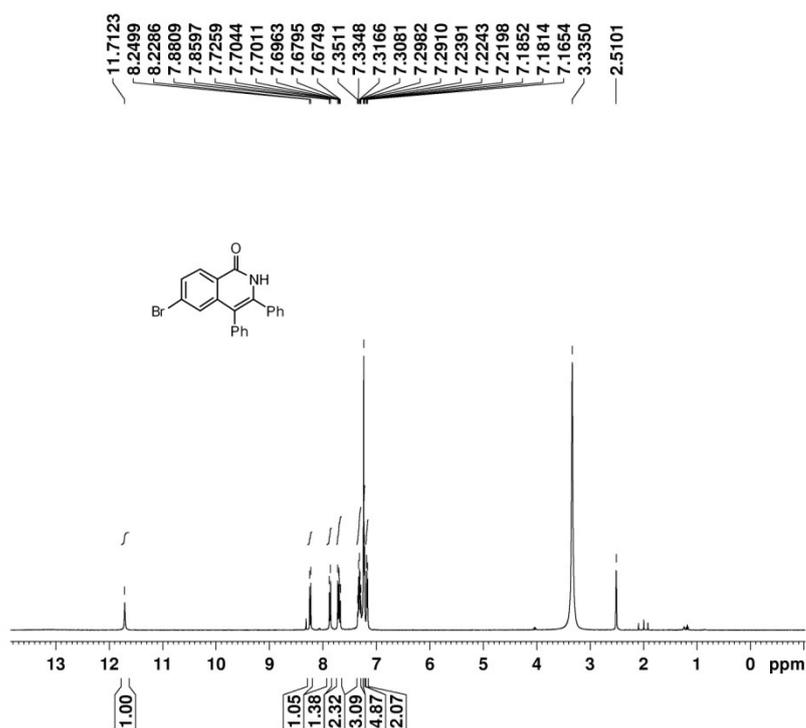
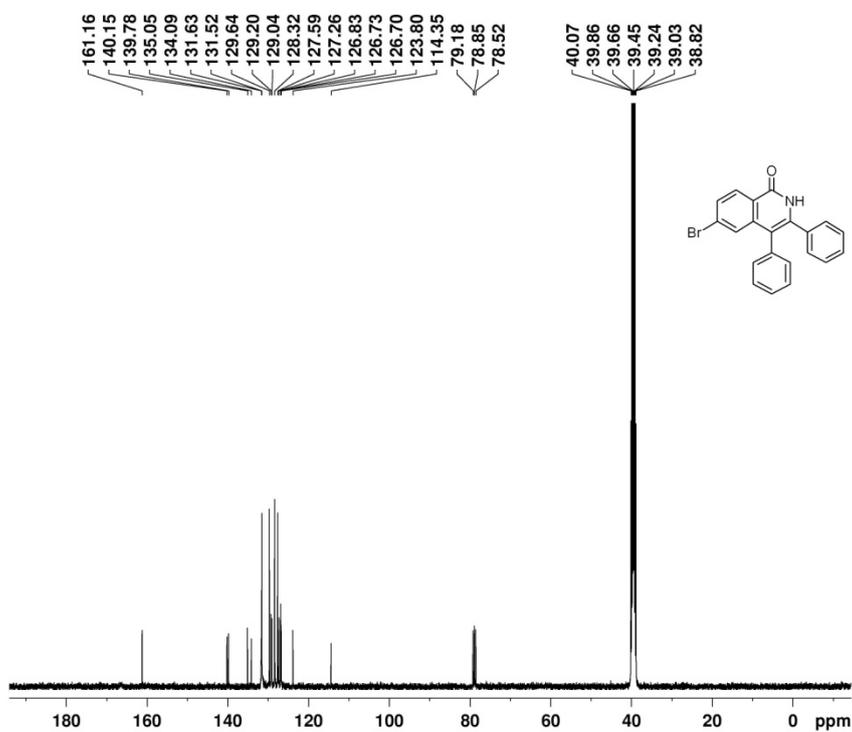
Current Data Parameters
NAME 22-04-2514.400 AN
EXPNO 610
PROCNO 1

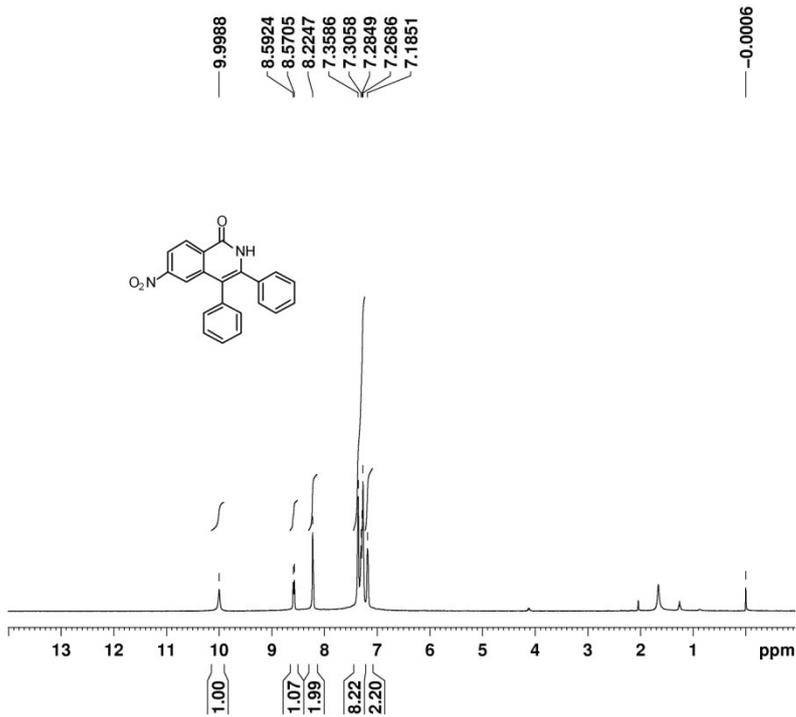
F2 - Acquisition Parameters
DATE_ 2016022
TIME 25.42
INSTRUM spect
PROBHD 5 mm PABBO 50/
PULPROG zgpg30
ID 65516
SOLVENT DMF
NS 2
DS 2
SWH 8012.600 Hz
FIDRES 0.122266 Hz
AQ 4.097465 sec
RG 14.26
AQ 57.400 usec
DE 1.00 usec
TE 300.2 K
SI 1.0000000 sec
TOP

===== CHANNEL f1 =====
NUC1 101.6261200 MHz
P1 12.10 usec
PL1 0.0000000 dB
PLM1 14.0000000 dB

F2 - Processing parameters
SI 65516
SF 400.1430000 MHz
WDW EM
SSB 0
GB 0
PC 1.00
  
```

Fig. 52: <sup>13</sup>C NMR of 4fa

Fig. 53:  $^1\text{H}$  NMR of 4gaFig. 54:  $^{13}\text{C}$  NMR of 4ga



RKBK-448

```

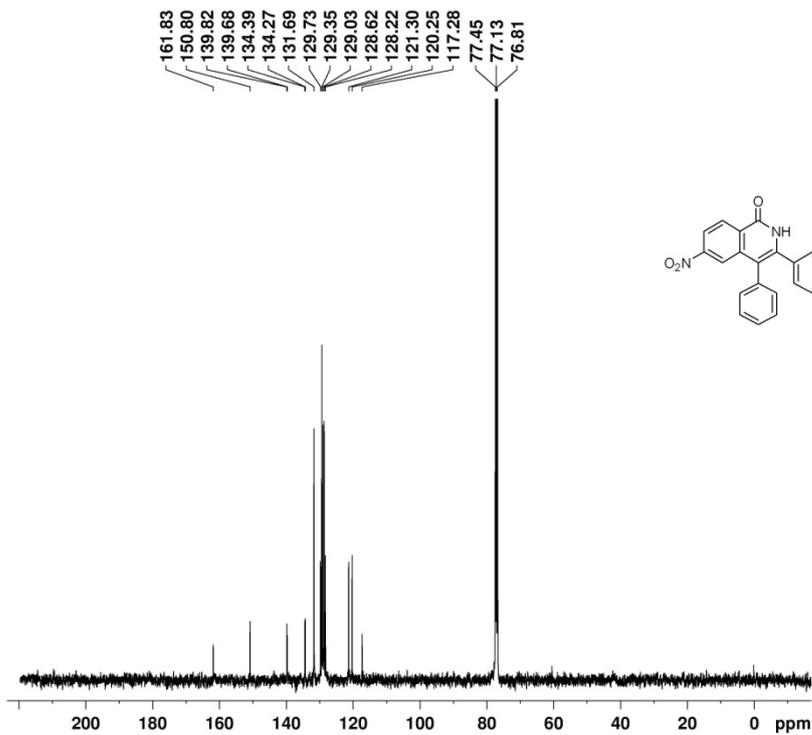
Current Data Parameters
NAME      23-04-14_10
EXPNO    650
PROCNO   1

F2 - Acquisition Parameters
Date_    20140429
Time     16.27
INSTRUM  spect
PROBHD   5 mm PABBO 501
PULPROG  zg30
TD        65536
SOLVENT  CDCl3
NS        8
DS        0
SWH       8012.820 Hz
FIDRES   0.122366 Hz
AQ        4.0894465 sec
RG        129.57
DM        62.400 usec
DE        6.50 usec
TE        300.0 K
D1        1.0000000 sec
TD0       1

----- CHANNEL f1 -----
SFO1     400.1629712 MHz
NUC1      1H
P1        12.35 usec
PLW1     14.0000000 W

F2 - Processing parameters
SI        65536
SF        400.1605997 MHz
WDW       EM
SSB       0
LB        0.30 Hz
GB        0
PC        1.00
  
```

Fig. 55: <sup>1</sup>H NMR of 4ha



RKBK-448

```

Current Data Parameters
NAME      02-05-2014_400_13CFE
EXPNO    620
PROCNO   1

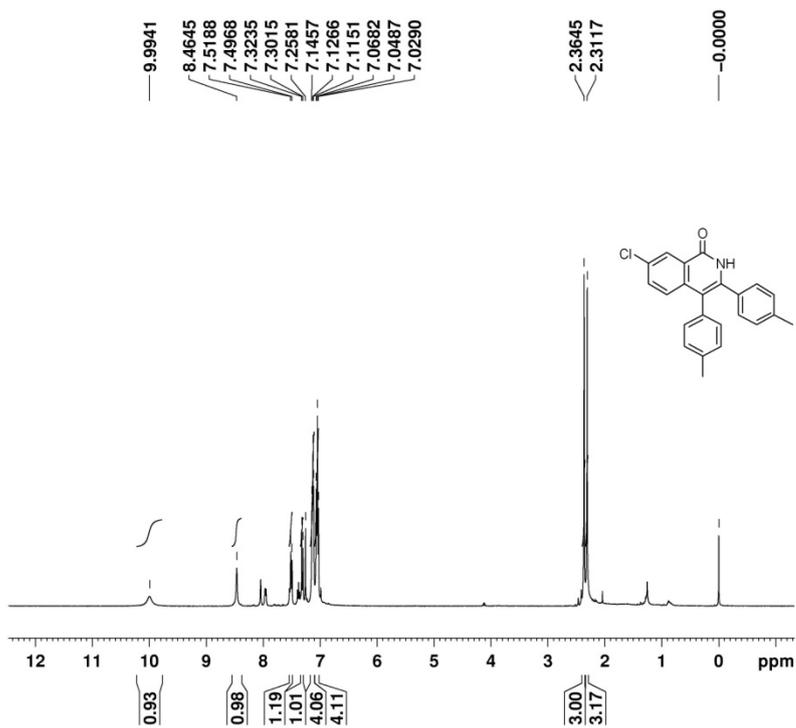
F2 - Acquisition Parameters
Date_    0140102
Time     21.44
INSTRUM  spect
PROBHD   5 mm PABBO 501
PULPROG  zgpg30
TD        65536
SOLVENT  CDCl3
NS        8
DS        0
SWH       24038.461 Hz
FIDRES   0.340798 Hz
AQ        1.362166 sec
RG        201.48
DM        20.800 usec
DE        6.50 usec
TE        300.0 K
D1        2.0000000 sec
D11       0.0300000 sec
TD0       1

----- CHANNEL f1 -----
SFO1     100.6104992 MHz
NUC1      13C
P1        9.00 usec
PLW1     61.8999987 W

----- CHANNEL f2 -----
SFO2     400.1629712 MHz
NUC2      1H
CQ13MG12 13
PCPD2    90.00 usec
P1MG12  13.1000000 W
PLW12    0.2670000 W
P1MG12   0.2163099 W

F2 - Processing parameters
SI        65536
SF        100.6204712 MHz
WDW       EM
SSB       0
LB        3.00 Hz
GB        0
PC        1.00
  
```

Fig. 56: <sup>13</sup>C NMR of 4ha



RKBK-446U

```

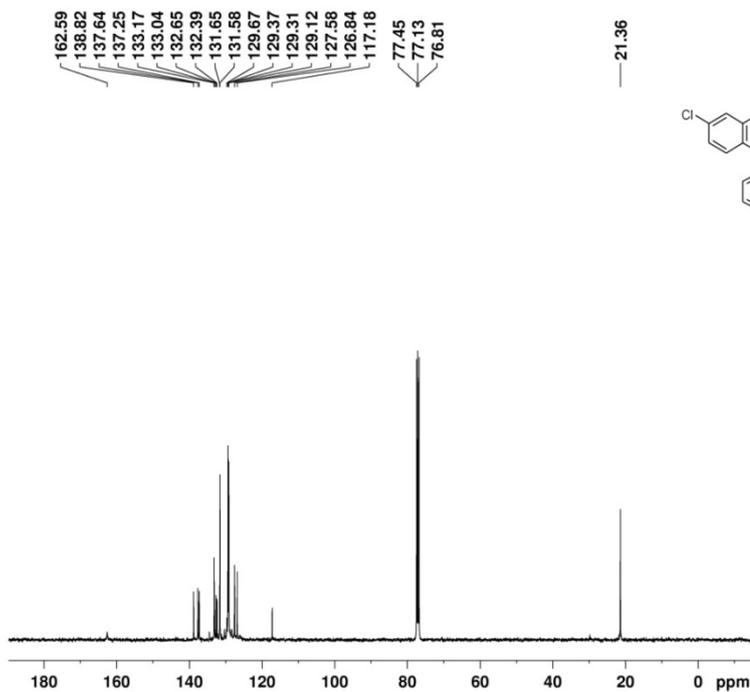
Current Data Parameters
NAME      25-04-14 AN 10
EXPNO    610
PROCNO   1

F2 - Acquisition Parameters
Date_    20160425
Time     16.18
INSTRUM  spect
PROBHD   5 mm PABBO BB/
PULPROG  zgpg30
TD       65536
SOLVENT  CDCl3
NS       8
DS       0
SWH      9615.385 Hz
FIDRES   0.144779 Hz
AQ       3.4078720 sec
RG       314.24
DW       52.000 usec
DE       6.50 usec
TS       300.0 K
D1       1.0000000 sec
TDO      1

===== CHANNEL f1 =====
SFO1     400.1429712 MHz
NUC1      13C
P1       12.35 usec
PL1      0.00000000 M

F2 - Processing parameters
SI       65536
SF       400.1400000 MHz
WDW      EM
SSB      0
LB       0.30 Hz
GB       0
PC       1.00
  
```

Fig. 57: <sup>1</sup>H NMR of 4ha



RKBK-446O

```

Current Data Parameters
NAME      25-04-14 AN 10
EXPNO    610
PROCNO   1

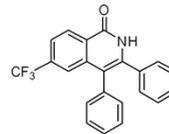
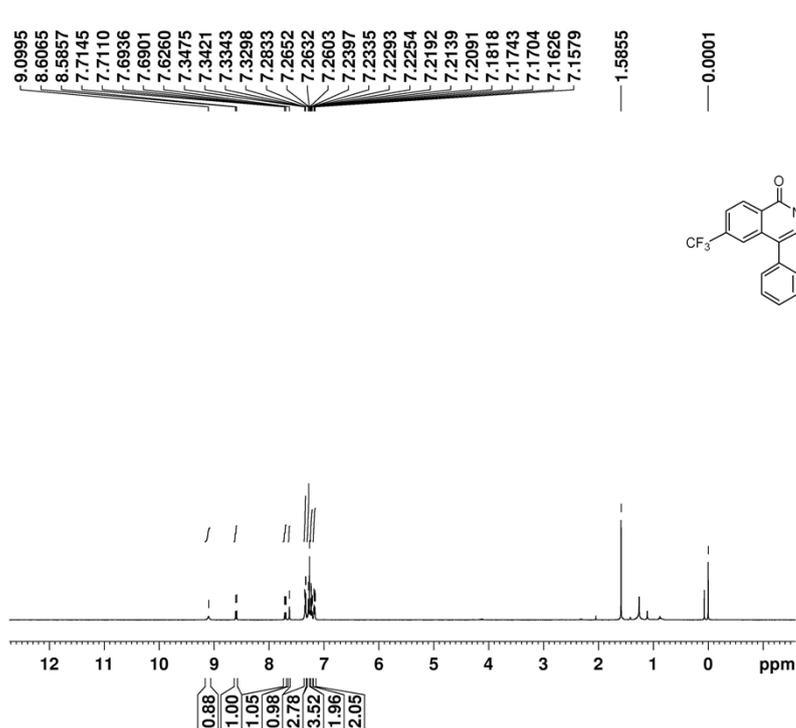
F2 - Acquisition Parameters
Date_    20160425
Time     16.18
INSTRUM  spect
PROBHD   5 mm PABBO BB/
PULPROG  zgpg30
TD       65536
SOLVENT  CDCl3
NS       8
DS       0
SWH      9615.385 Hz
FIDRES   0.144779 Hz
AQ       3.4078720 sec
RG       314.24
DW       52.000 usec
DE       6.50 usec
TS       300.0 K
D1       1.0000000 sec
TDO      1

===== CHANNEL f1 =====
SFO1     400.1429712 MHz
NUC1      13C
P1       12.35 usec
PL1      0.00000000 M

F2 - Processing parameters
SI       65536
SF       400.1400000 MHz
WDW      EM
SSB      0
LB       0.30 Hz
GB       0
PC       1.00
  
```

Fig. 58: <sup>13</sup>C NMR of 4ib





RKBK-439

```

Current Data Parameters
NAME 27-08-2014.400
EXPNO 620
PROCNO 1

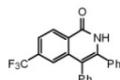
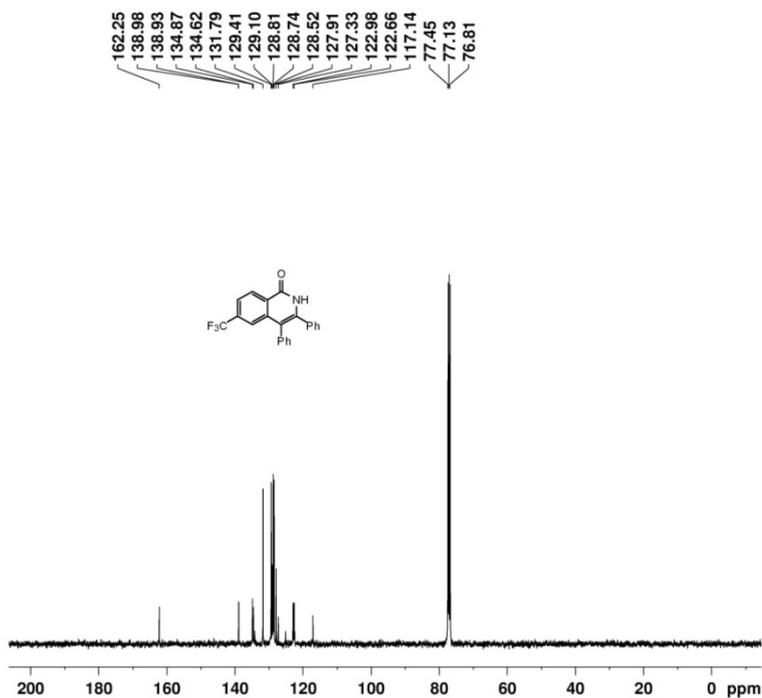
F2 - Acquisition Parameters
Date_ 20140827
Time 17.19
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zgpg30
TD 65536
SOLVENT CDCl3
NS 8
DS 0
SWH 9615.385 Hz
FIDRES 0.146719 Hz
AQ 3.4078720 sec
RG 159.22
DW 52.000 usec
DE 6.50 usec
TE 300.0 K
D1 1.0000000 sec
TD0 1

----- CHANNEL f1 -----
SFO1 400.1629712 MHz
NUC1 1H
P1 12.85 usec
PLW1 13.10000038 W

F2 - Processing parameters
SI 65536
SF 400.1600999 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.00

```

Fig. 61: <sup>1</sup>H NMR of 4ja



RKBK-439

```

Current Data Parameters
NAME 27-08-2014.400
EXPNO 620
PROCNO 1

F2 - Acquisition Parameters
Date_ 20140827
Time 17.19
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zgpg30
TD 65536
SOLVENT CDCl3
NS 8
DS 0
SWH 9615.385 Hz
FIDRES 0.146719 Hz
AQ 3.4078720 sec
RG 159.22
DW 52.000 usec
DE 6.50 usec
TE 300.0 K
D1 1.0000000 sec
TD0 1

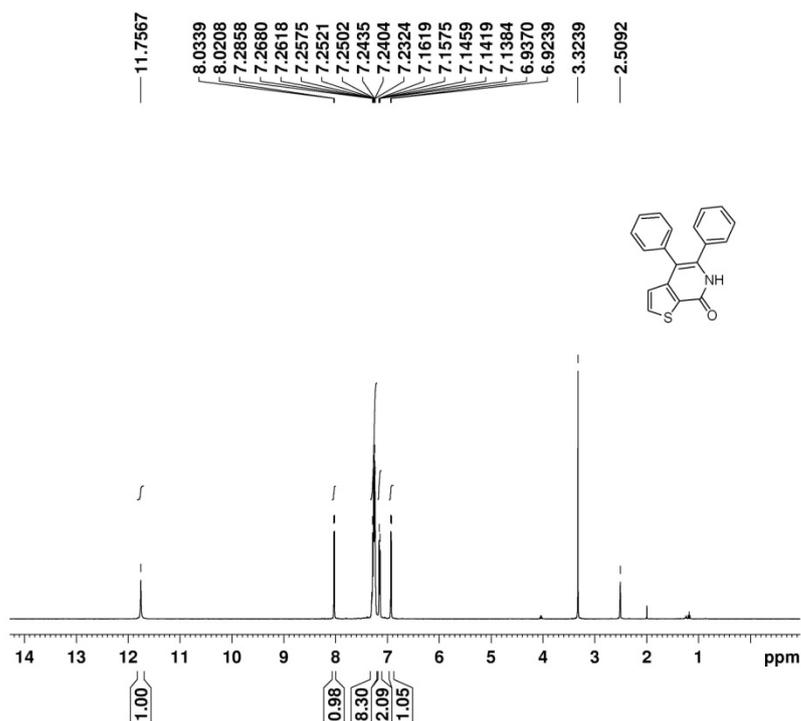
----- CHANNEL f1 -----
SFO1 400.1629712 MHz
NUC1 1H
P1 12.85 usec
PLW1 13.10000038 W

F2 - Processing parameters
SI 65536
SF 400.1600999 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.00

```

Fig. 62: <sup>13</sup>C NMR of 4ja

RKKB-443



```

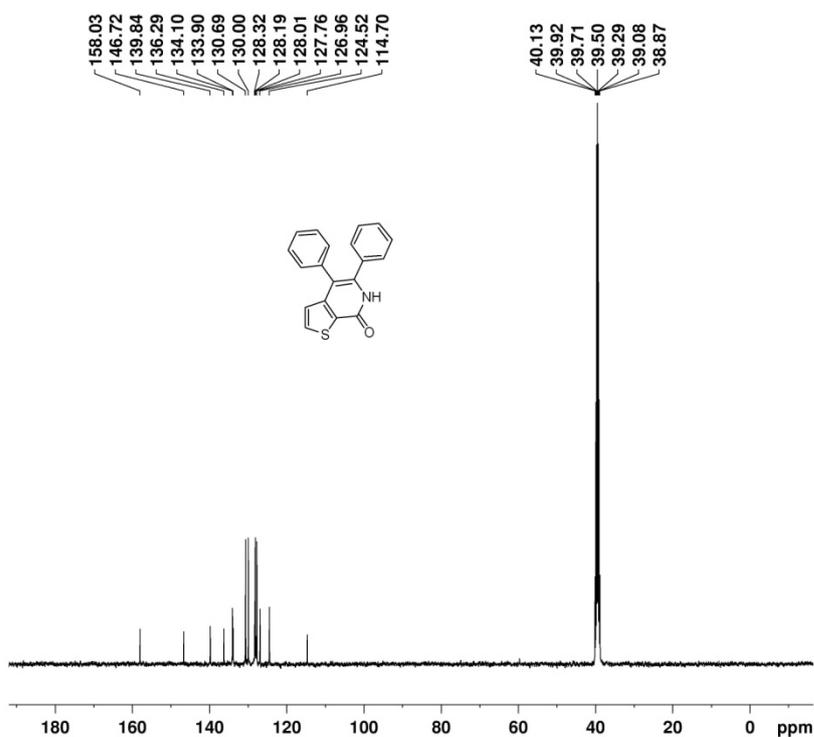
Current Data Parameters
NAME      22-04-2014_400 AN
EXPNO    330
PROCNO   1

F2 - Acquisition Parameters
Date_    20140422
Time     20.50
INSTRUM  spect
PROBHD   5 mm PABBO 50/
PULPROG  zg30
TD        65536
SOLVENT  DMSO
NS        8
DS        2
SWH       8012.820 Hz
FIDRES    0.122246 Hz
AQ         4.0894455 sec
RG         114.26
DW         62.400 usec
DE         6.50 usec
TE        300.0 K
D1         1.0000000 sec
TD0        1

===== CHANNEL f1 =====
SFO1     400.1429712 MHz
NUC1      1H
P1        12.35 usec
PL1       0.00000000 W
F1W1      14.00000000 W

F2 - Processing parameters
SI        65536
SF        400.1605000 MHz
WDW       EM
SSB       0
LB        0.30 Hz
GB        0
PC        1.00
  
```

Fig. 63: <sup>1</sup>H NMR of 4ka



RKKB-443

```

Current Data Parameters
NAME      22-04-2014_400-13C AN
EXPNO    330
PROCNO   1

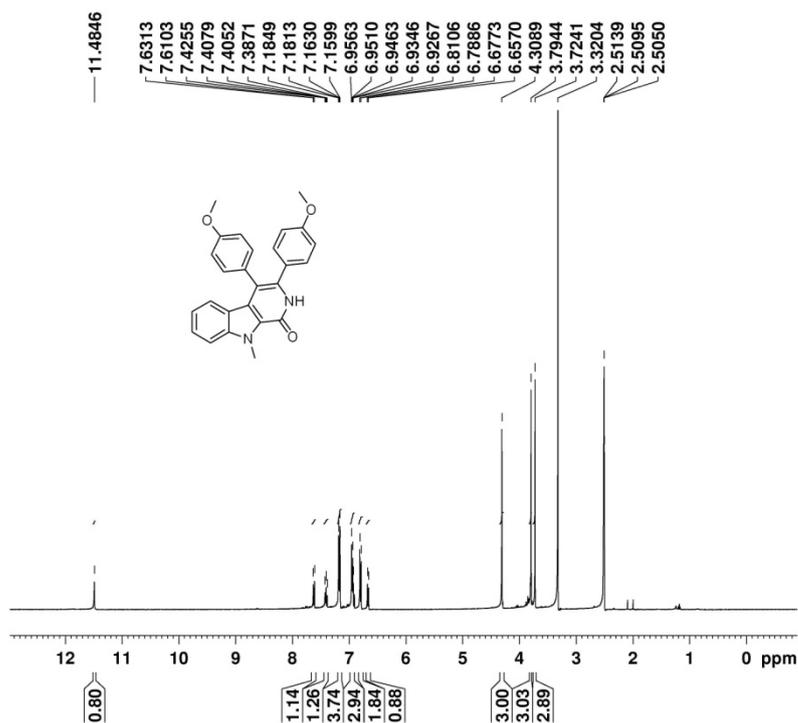
F2 - Acquisition Parameters
Date_    20140422
Time     20.50
INSTRUM  spect
PROBHD   5 mm PABBO 50/
PULPROG  zgpg30
TD        65536
SOLVENT  DMSO
NS        8
DS        2
SWH       125.76000 MHz
FIDRES    0.122246 Hz
AQ         4.0894455 sec
RG         114.26
DW         62.400 usec
DE         6.50 usec
TE        300.0 K
D1         1.0000000 sec
TD0        1

===== CHANNEL f1 =====
SFO1     125.7600000 MHz
NUC1      13C
P1        12.35 usec
PL1       0.00000000 W
F1W1      14.00000000 W

F2 - Processing parameters
SI        65536
SF        125.7600000 MHz
WDW       EM
SSB       0
LB        0.30 Hz
GB        0
PC        1.00
  
```

Fig. 64: <sup>13</sup>C NMR of 4ka

RKKB-423



```

Current Data Parameters
NAME 02-04-2014_403 FN
EXPNO 630
PROCNO 1

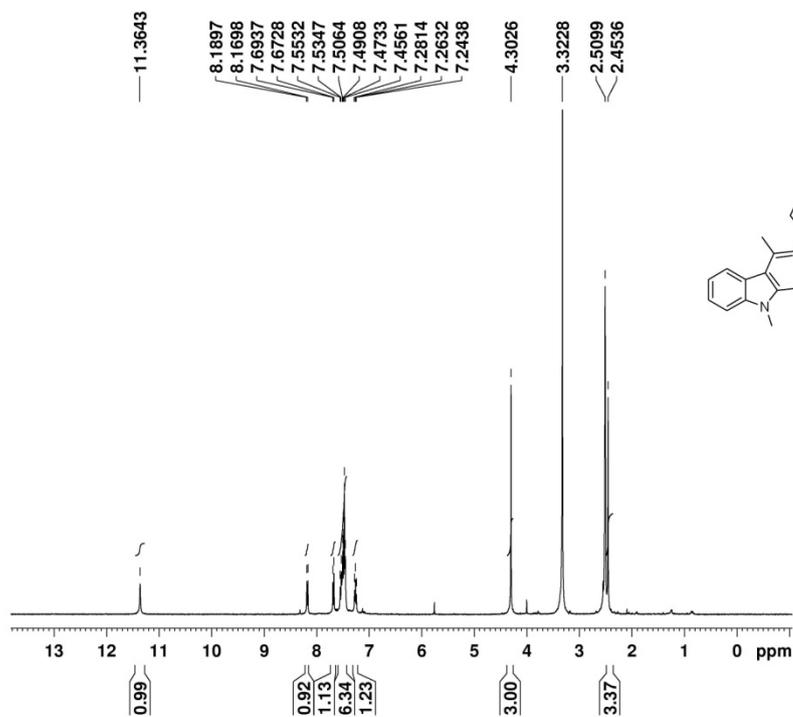
F2 - Acquisition Parameters
Date_ 20140402
Time 17.19
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zgpg30
TD 65536
SOLVENT DMSO
NS 16
DS 0
SWH 8012.820 Hz
FIDRES 0.122266 Hz
AQ 4.0394665 sec
RG 159.22
DM 62.400 usec
DE 6.30 usec
TE 300.2 K
D1 1.0000000 sec
TD0 1

----- CHANNEL f1 -----
NUC1 400.1623712 MHz
P1 12.85 usec
PLM1 13.10000000 W

F2 - Processing parameters
SI 65536
SF 400.1600000 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.00
  
```

Fig. 65: <sup>1</sup>H NMR of 4lc

RKKB-426



```

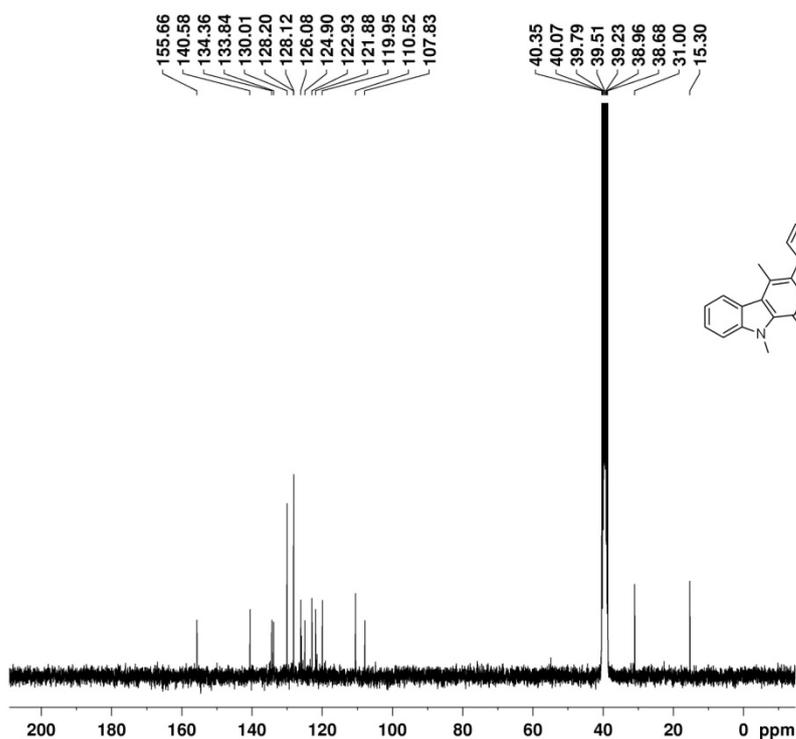
Current Data Parameters
NAME 03-04-14 FN 10
EXPNO 607
PROCNO 1

F2 - Acquisition Parameters
Date_ 20140304
Time 15.15
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zgpg30
TD 65536
SOLVENT DMSO
NS 16
DS 0
SWH 8012.820 Hz
FIDRES 0.122266 Hz
AQ 4.0394665 sec
RG 159.22
DM 62.400 usec
DE 6.30 usec
TE 300.2 K
D1 1.0000000 sec
TD0 1

----- CHANNEL f1 -----
NUC1 400.1623712 MHz
P1 12.85 usec
PLM1 13.10000000 W

F2 - Processing parameters
SI 65536
SF 400.1600000 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.00
  
```

Fig. 66: <sup>1</sup>H NMR of 4lg

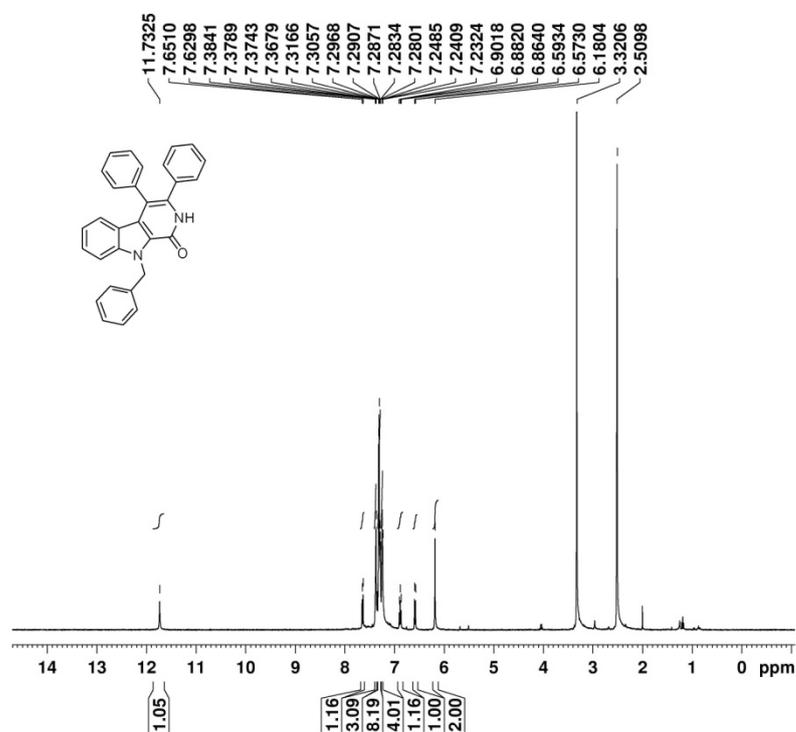


RKBK-C10

```

Current Data Parameters
NAME 13-06-14 FR 18
EXPNO 1
PROCNO 1
F2 - Processing parameters
SI 1
SF 50.67784 MHz
WDW 0
SSB 0
LB 1.00 Hz
GB 0
PC 1.40
  
```

Fig. 67: <sup>13</sup>C NMR of 4lg

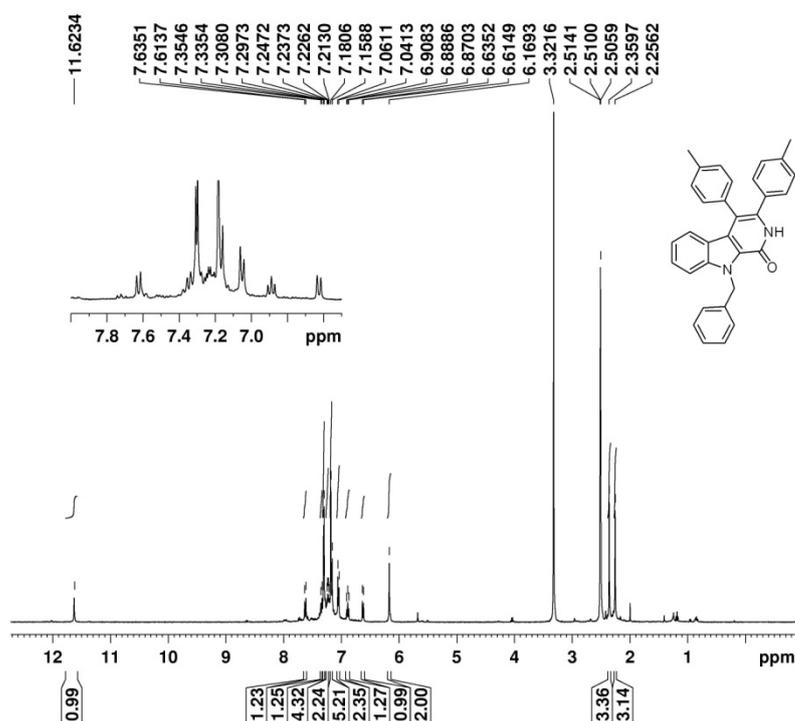


RKBK-431

```

Current Data Parameters
NAME 04-04-14 FR 18
EXPNO 1
PROCNO 1
F2 - Acquisition Parameters
Date_ 20140404
Time 15.51
INSTRUM spect
PROBHD 5 mm PABBO BBI
PULPROG zgpg30
TD 65536
SOLVENT DMSO
NS 8
DS 0
SWH 8012.820 Hz
FIDRES 0.122266 Hz
AQ 4.0894465 sec
RG 159.22
DM 62.400 usec
DE 6.50 usec
TE 300.2 K
D1 1.0000000 sec
TDG 1
----- CHANNEL f1 -----
SFO1 400.1429712 MHz
NUC1 13
P1 12.85 usec
PL1 13.10000000 W
F2 - Processing parameters
SI 1
SF 400.1430000 MHz
WDW 0
SSB 0
LB 0.30 Hz
GB 0
PC 1.00
  
```

Fig. 68: <sup>1</sup>H NMR of 4ma



RKBK-430

```

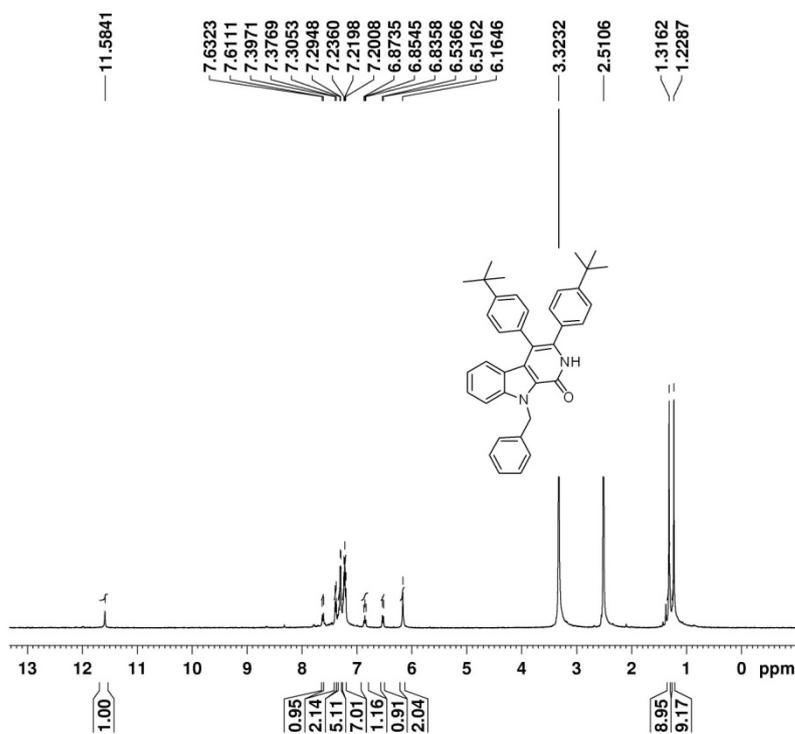
Current Data Parameters
NAME 04-04-14 FN 1H
EXPNO 610
PROCNO 1

F2 - Acquisition Parameters
Date_ 20140404
Time 15.47
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zg30
TD 65536
SOLVENT DMSO
NS 8
DS 0
SWH 8012.820 Hz
FIDRES 0.122266 Hz
AQ 4.0894465 sec
RG 159.22
DW 62.400 usec
DE 6.50 usec
TE 300.0 K
DL 1.0000000 sec
TDO 1

===== CHANNEL f1 =====
SFO1 400.1629712 MHz
NUC1 1H
P1 12.89 usec
PLW1 13.10000038 W

F2 - Processing parameters
SI 65536
SF 400.1605000 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.00
  
```

Fig. 69: <sup>1</sup>H NMR of 4mb



RKBK-428

```

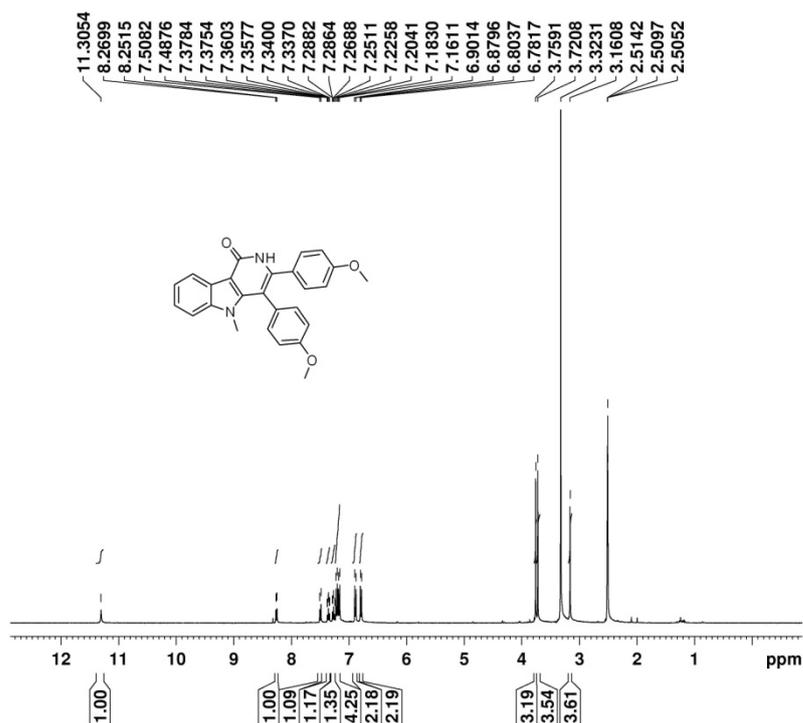
Current Data Parameters
NAME 03-04-14 FN 1H
EXPNO 640
PROCNO 1

F2 - Acquisition Parameters
Date_ 20140403
Time 15.33
INSTRUM spect
PROBHD 5 mm PABBO BB/
PULPROG zg30
TD 65536
SOLVENT DMSO
NS 8
DS 0
SWH 8012.820 Hz
FIDRES 0.122266 Hz
AQ 4.0894465 sec
RG 145.29
DW 62.400 usec
DE 6.50 usec
TE 300.0 K
DL 1.0000000 sec
TDO 1

===== CHANNEL f1 =====
SFO1 400.1629712 MHz
NUC1 1H
P1 12.85 usec
PLW1 13.10000038 W

F2 - Processing parameters
SI 65536
SF 400.1605000 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.00
  
```

Fig. 70: <sup>1</sup>H NMR of 4md

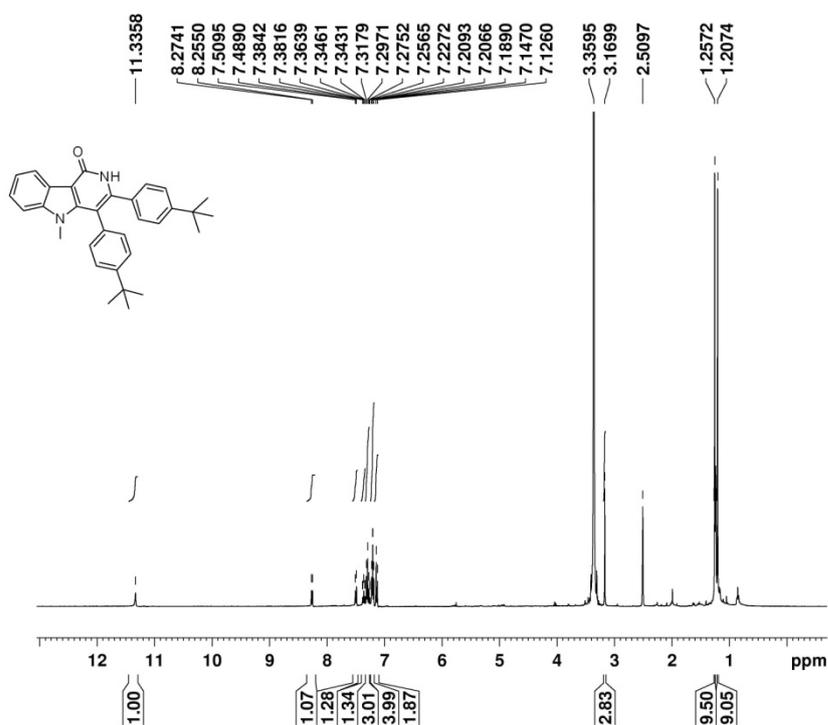


RKBK-429

```

Current Data Parameters
NAME      03-04-14 F9 10
EXPNO    630
PROCNO   1
----- Acquisition Parameters
Date_    20140405
Time     15.37
INSTRUM  spect
PROBHD   5 mm PABBO QNP
PULPROG  zg30
TD        65536
SOLVENT  DMSO
DS        2
SWH       8012.820 Hz
FIDRES   0.122266 Hz
AQ        4.0894465 sec
RG        119.2
WDW        62.400 usec
SSB        0
TE        300.2 K
D1        1.00000000 sec
TD0       1
----- CHANNEL f1 -----
SFO1     400.1429712 MHz
NUC1     13
P1        12.80 usec
P1M1     13.10000000 W
----- Processing parameters
SI        65536
SF        400.1400000 MHz
WDW       62.400 usec
SSB       0
GB        0
PC        1.00
  
```

Fig. 71: <sup>1</sup>H NMR of 4nc



RKBK-C1

```

Current Data Parameters
NAME      23-04-2014-400 AN
EXPNO    610
PROCNO   1
----- Acquisition Parameters
Date_    20140423
Time     14.22
INSTRUM  spect
PROBHD   5 mm PABBO QNP
PULPROG  zg30
TD        65536
SOLVENT  DMSO
DS        2
SWH       8012.820 Hz
FIDRES   0.122266 Hz
AQ        4.0894465 sec
RG        89.0
WDW        62.400 usec
SSB        0
TE        300.2 K
D1        1.00000000 sec
TD0       1
----- CHANNEL f1 -----
SFO1     400.1429712 MHz
NUC1     13
P1        12.80 usec
P1M1     14.00000000 W
----- Processing parameters
SI        65536
SF        400.1600000 MHz
WDW       62.400 usec
SSB       0
GB        0
PC        1.00
  
```

Fig. 72: <sup>1</sup>H NMR of 4nd

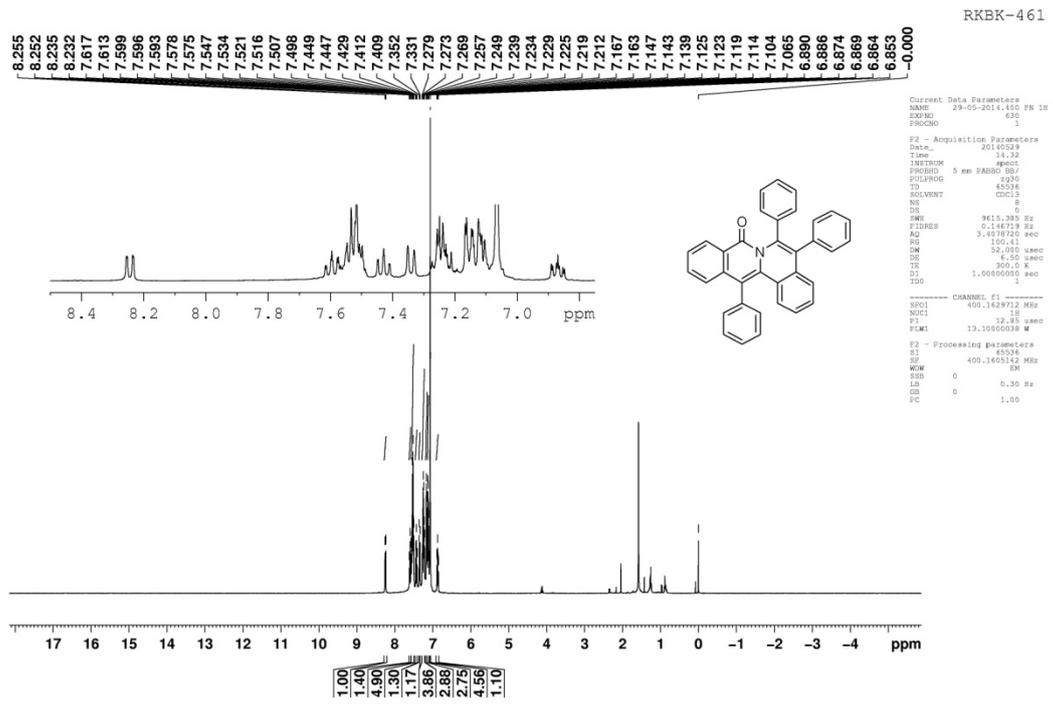


Fig. 73: <sup>1</sup>H NMR of 6

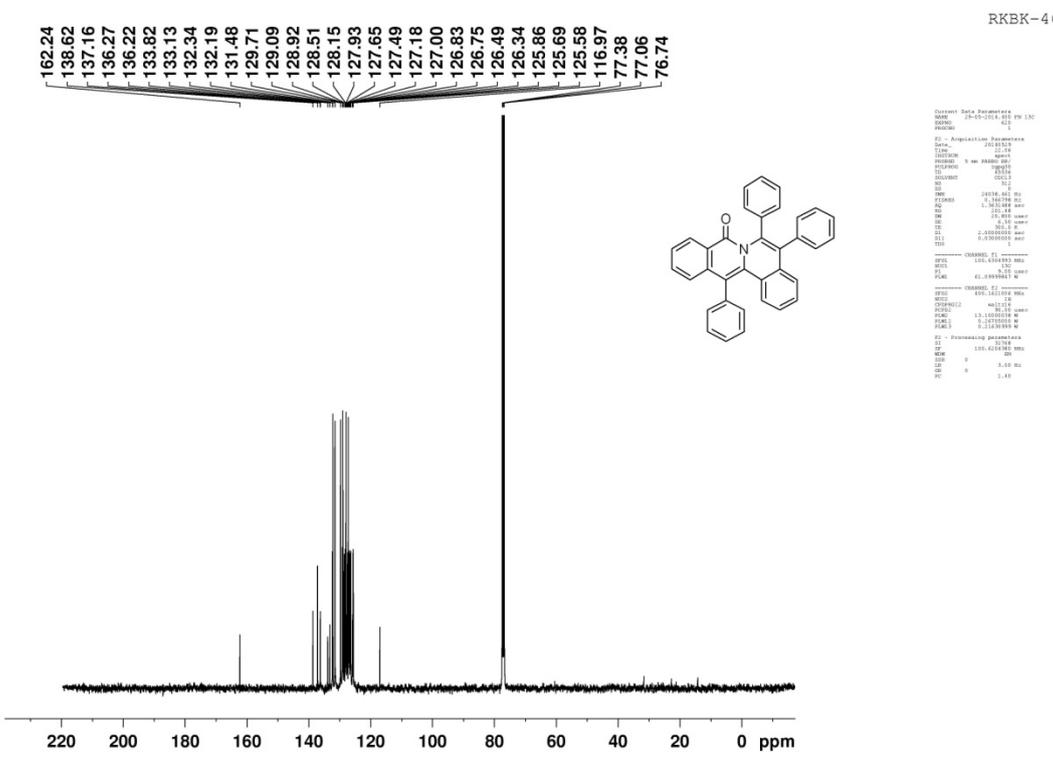


Fig. 74: <sup>13</sup>C NMR of 6

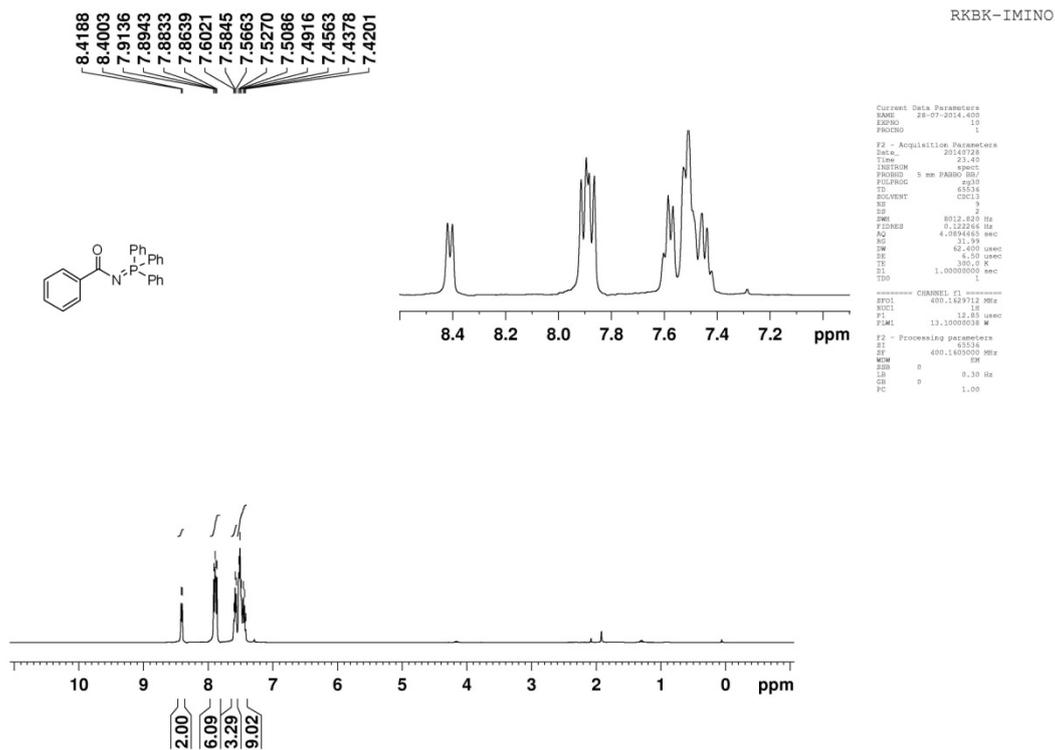
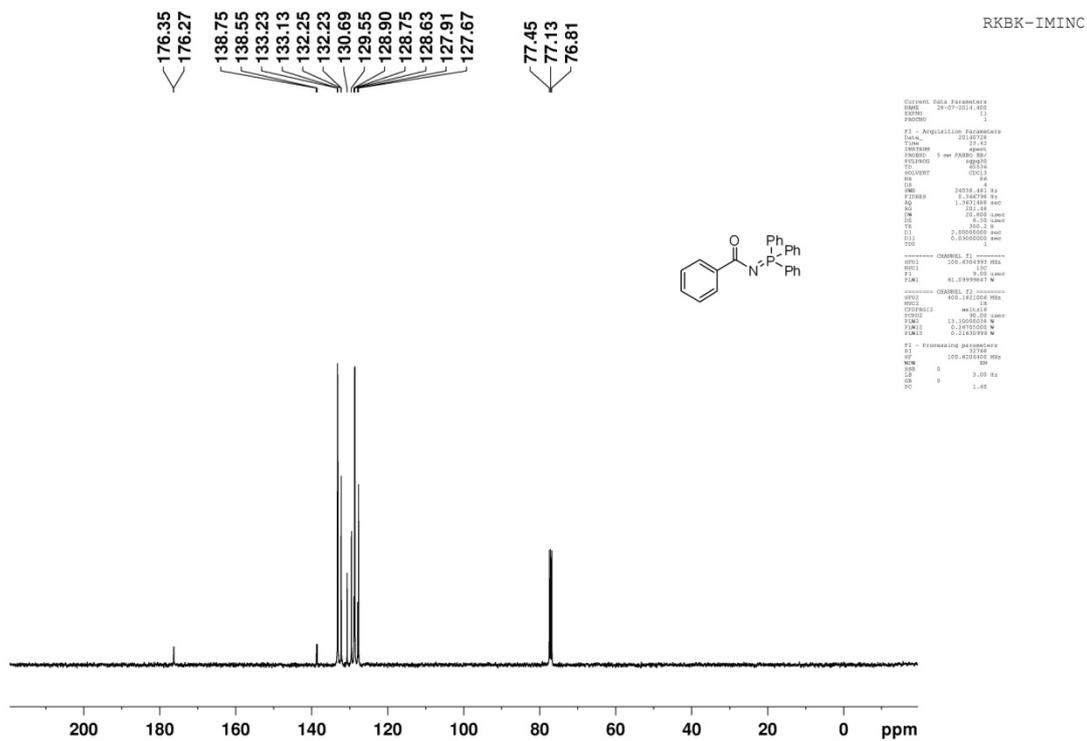


Fig. 75: <sup>1</sup>H NMR of 2a



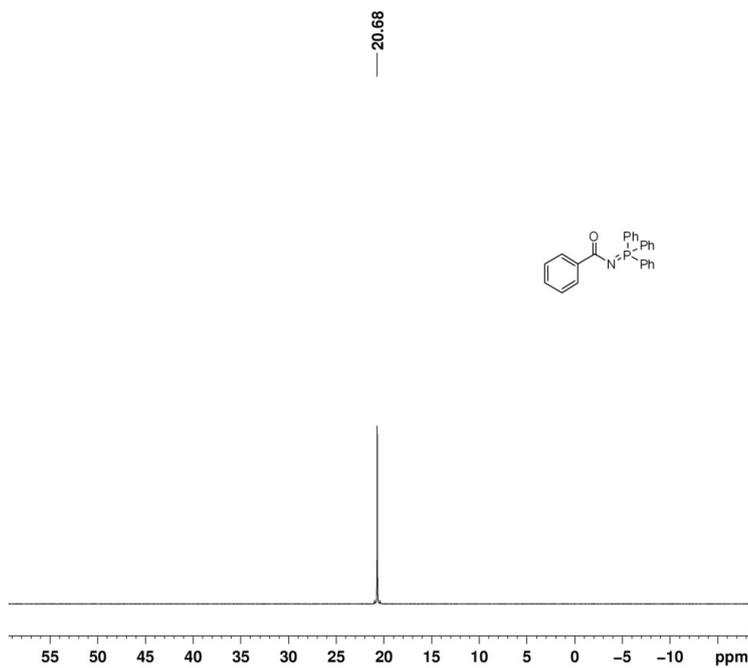


Fig. 77:  $^{31}\text{P}$  NMR of 2a

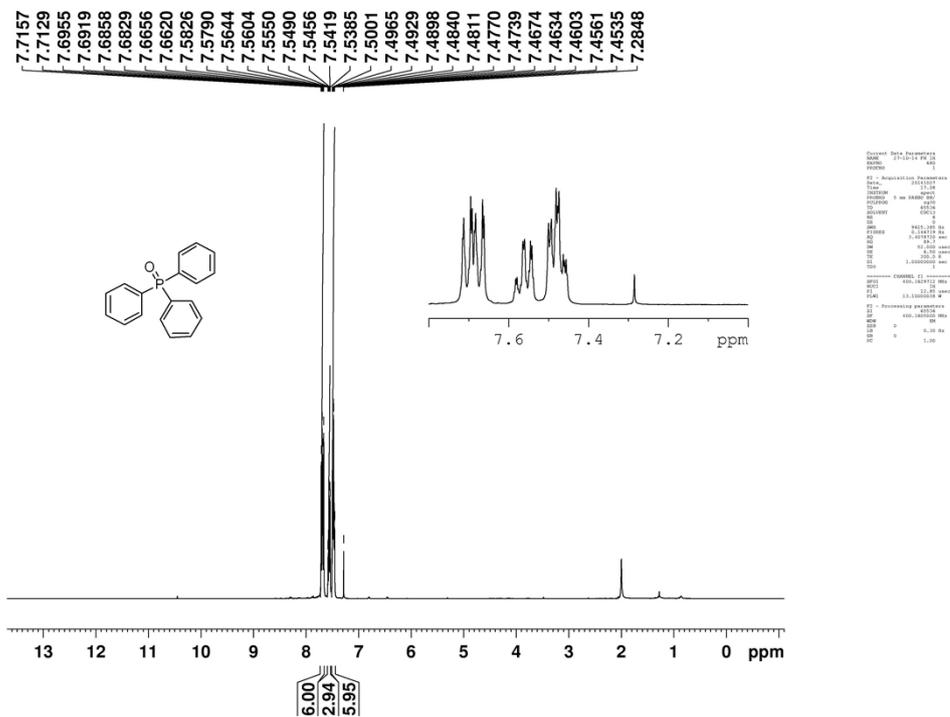
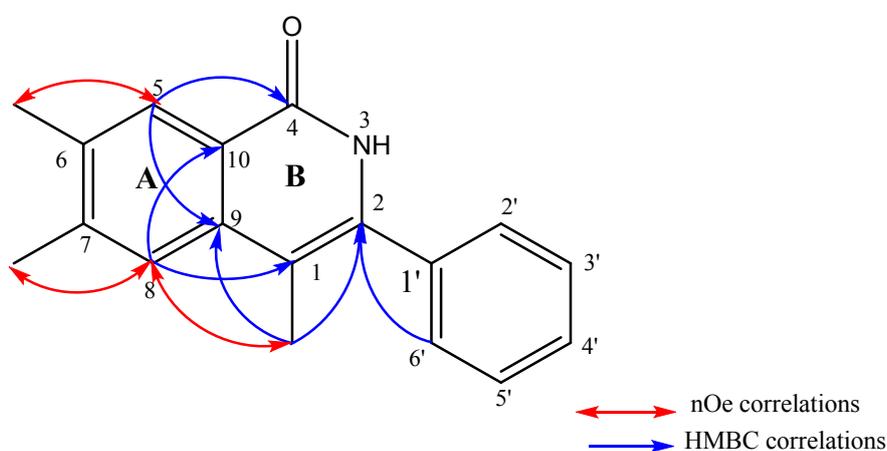
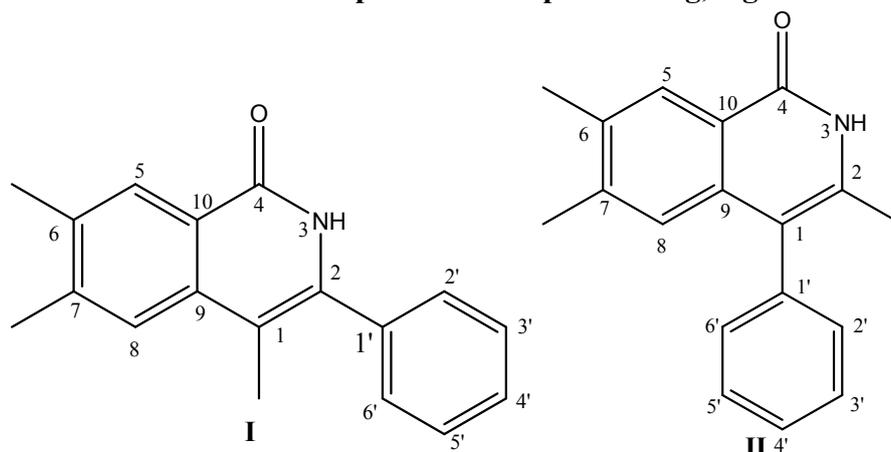


Fig. 78:  $^1\text{H}$  NMR of Triphenyl phosphine oxide



### VIII. 2-D spectra of compounds 4bg, 4lg

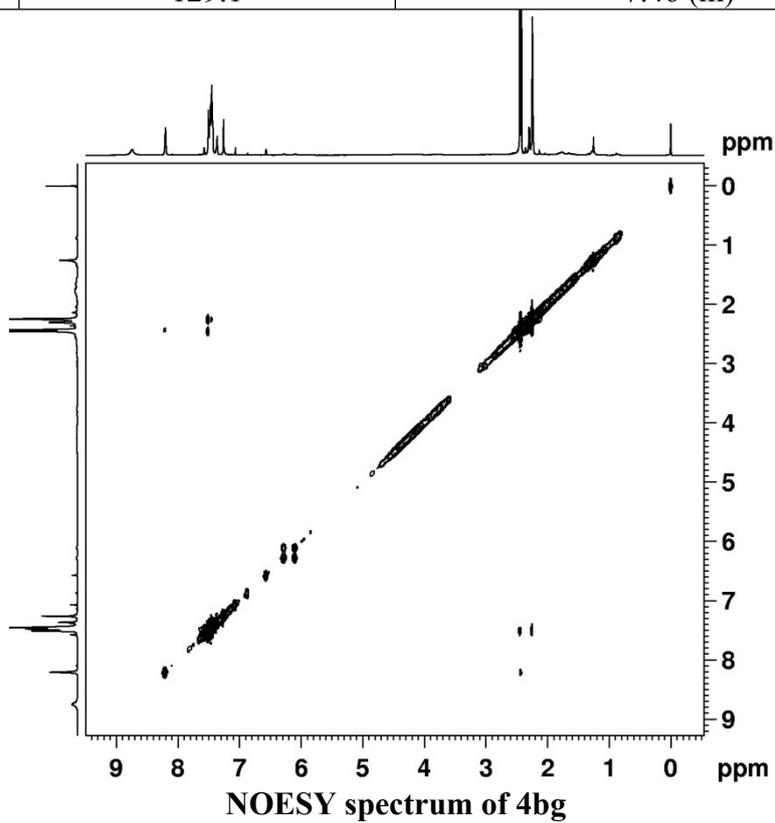


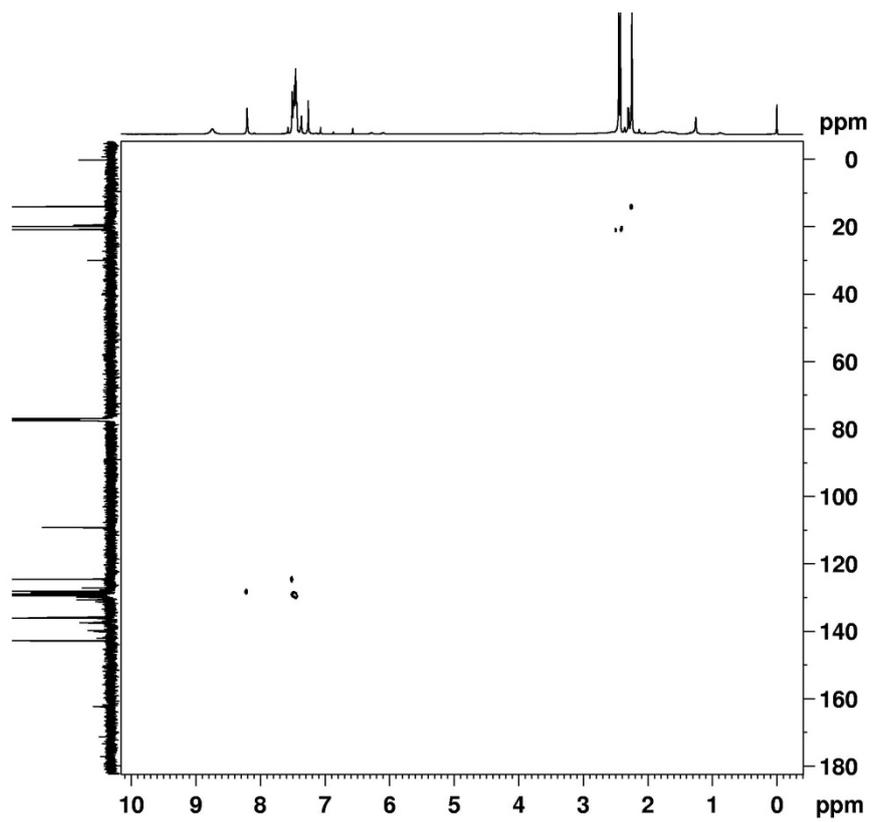
#### Important HMBC and nOe correlations of 4bg

The entire  $^1\text{H}$  and  $^{13}\text{C}$  NMR signal assignments and skeletal connectivity were established from a combination of various 1D and 2D NMR experiments. HMBC gave the information about the connectivity of atoms in the molecule while NOESY established the spatial correlations between proximate protons in the molecule. HMBC correlations of H-8 to C-1 and 1-CH<sub>3</sub> to C-9 indicated that CH<sub>3</sub> group is attached to C-1 in molecular structure I, whereas such HMBC correlations are not possible in molecular structure II. At the same time H-5 is giving HMBC correlation with quaternary carbon (C=O) C-4 and H-2',6' is also giving HMBC correlation with C-2, these correlations determined the connectivity of ring A to ring B and phenyl ring to ring B respectively. Important nOe correlation between 1-CH<sub>3</sub> and H-8 showed the proximity of both protons in the space and provided strong evidence for the attachment of CH<sub>3</sub> at C-1 atom.

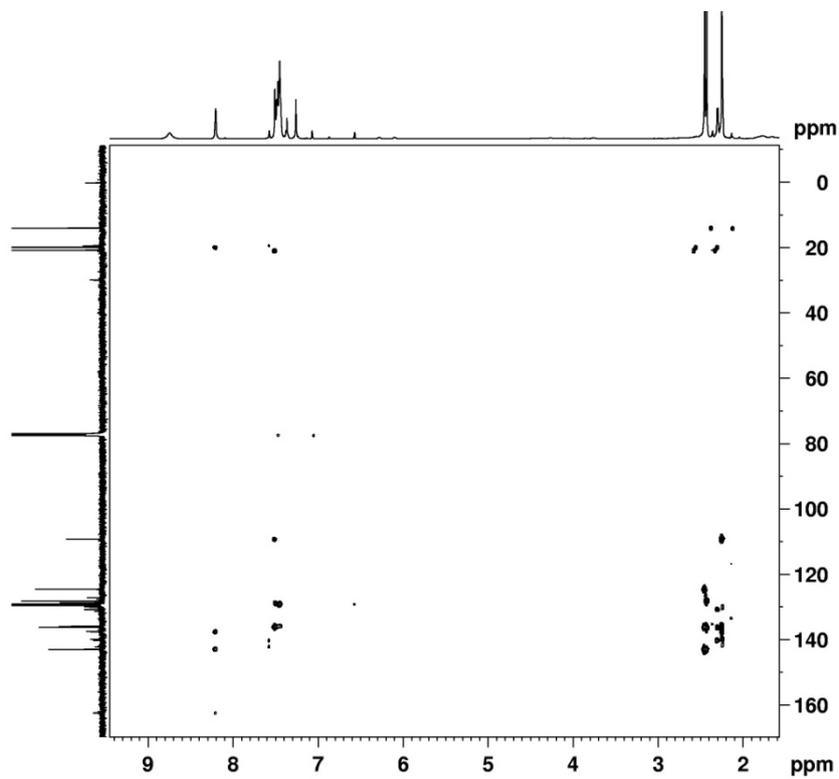
#### $^1\text{H}$ and $^{13}\text{C}$ signal assignments of 4bg

Carbon position	<sup>13</sup> C Chemical shift (ppm)	<sup>1</sup> H Chemical shift (ppm)
C-1	106.1	-
C-1-CH <sub>3</sub>	14.0	2.24 (s)
C-2	134.9	-
N-H-3	-	8.74 (s)
C-4	161.6	-
C-5	128.1	8.20 (s)
C-6	136.1	-
C-6-CH <sub>3</sub>	19.8	2.42 (s)
C-7	142.8	-
C-7-CH <sub>3</sub>	20.8	2.45 (s)
C-8	124.5	7.51 (s)
C-9	136.8	-
C-10	135.5	-
C-1'	136.4	-
C-2',C-6'	128.9	7.47 (m)
C-3',C-5'	129.4	7.45 (m)
C-4'	129.1	7.46 (m)



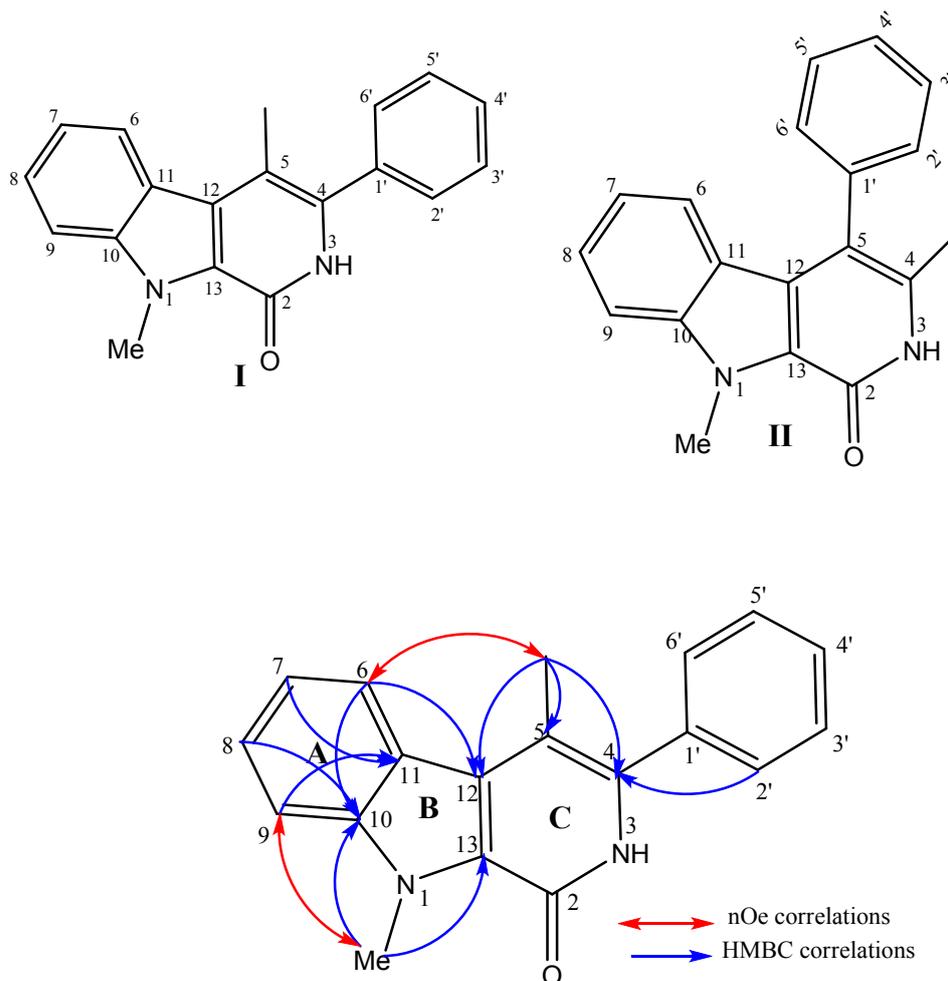


HSQC spectrum of 4bg



HMBC spectrum of 4bg

Compound-4lg



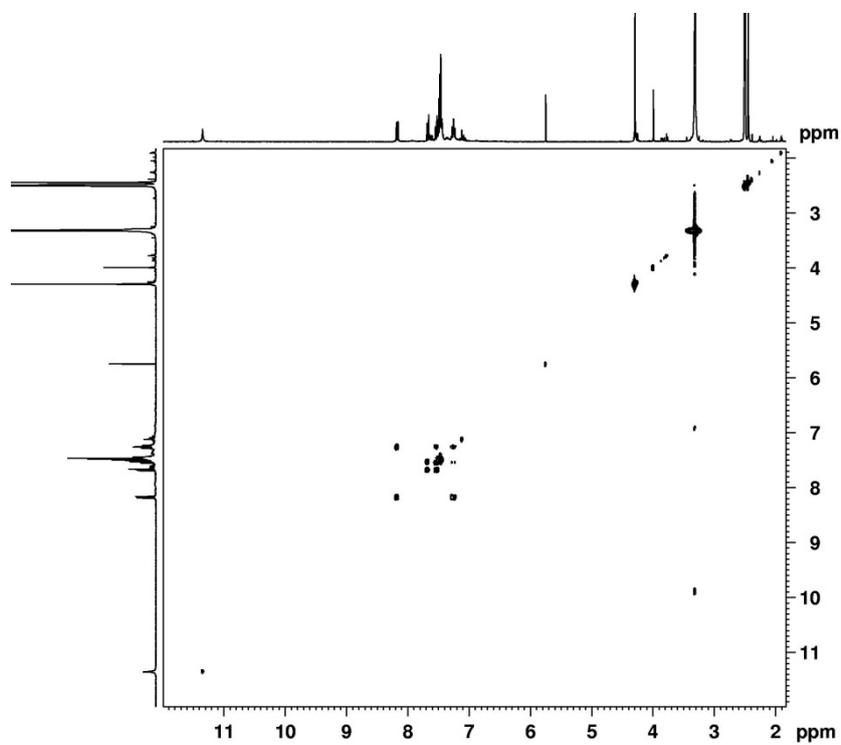
### Important HMBC and nOe correlations of 4lg

COSY experiment established the two spin systems H-6 to H-9 in ring C and phenyl ring. HMBC correlations of H-6 and 5-CH<sub>3</sub> to C-12 and 5-CH<sub>3</sub>, H-2', H-6' to C-4 clearly indicated that CH<sub>3</sub> group is attached to C-5 of ring C and phenyl ring to C-4 atoms respectively in molecular structure I, which is not possible in molecular structure II. At the same time H-6 and H-8 giving HMBC correlation to quaternary carbon C-10 and H-7 and H-9 is giving HMBC correlation with quaternary carbon C-11, and fixed the positions of quaternary carbons. Important nOe correlation between 5-CH<sub>3</sub> and H-6 showed the proximity of both protons in the space and played an important role to establish the attachment of methyl group at C-5 atom in structure I, such nOe correlation would be absent in structure II.

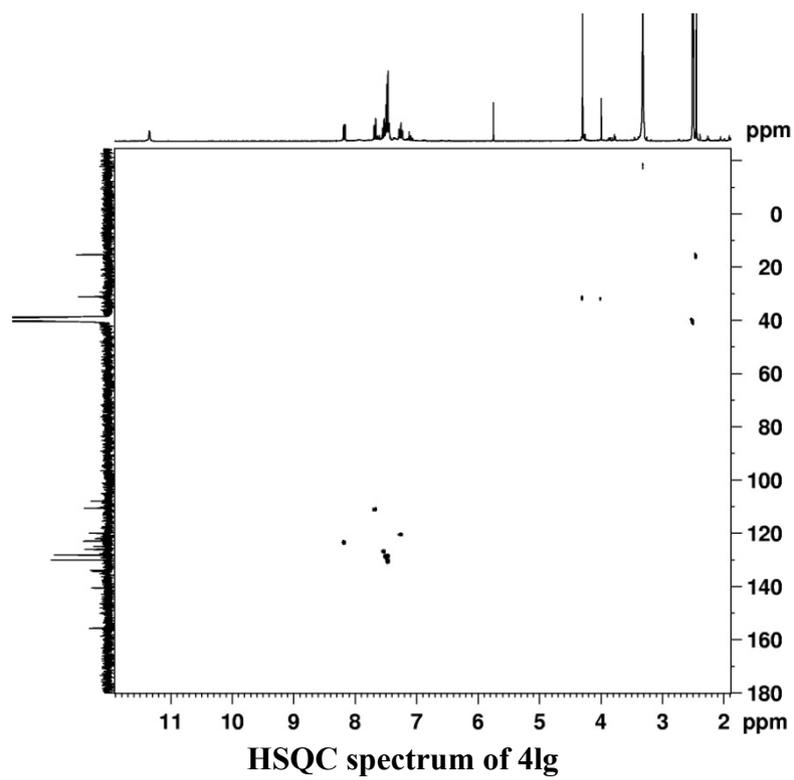
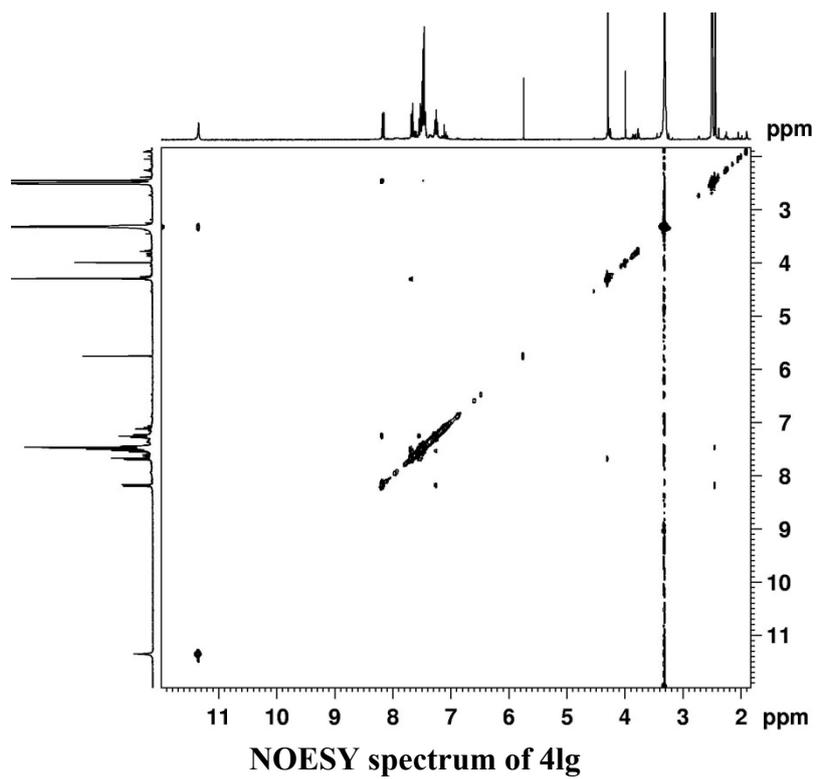
Carbon position	<sup>13</sup> C Chemical shift (ppm)	<sup>1</sup> H Chemical shift (ppm)
N-CH <sub>3</sub>	31.5	4.30 (s)
C-2	155.6	-
N-H-3	-	11.34 (s)
C-4	133.9	-
C-5	107.9	-

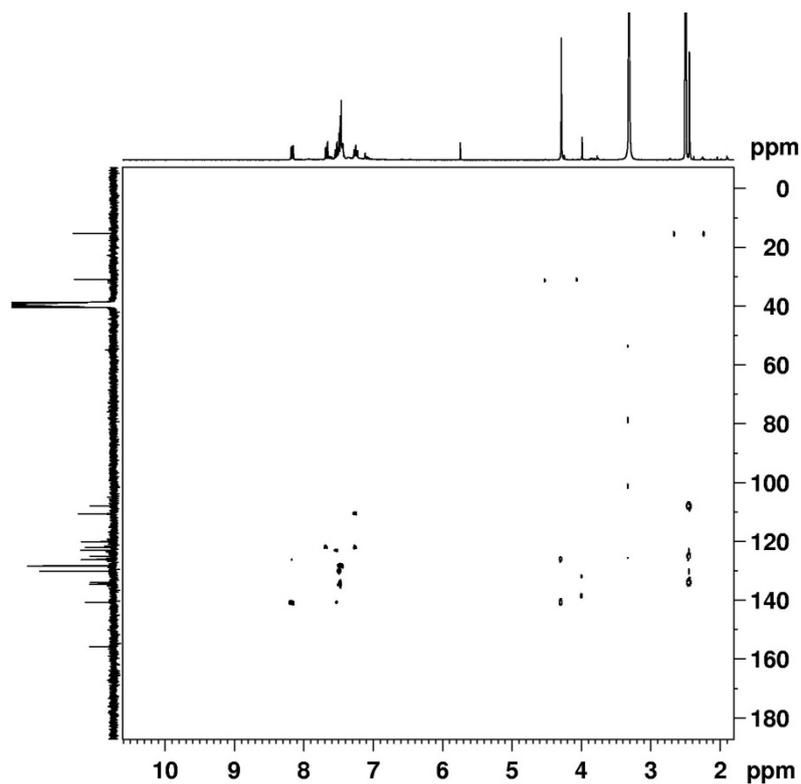
C-5-CH <sub>3</sub>	15.6	2.44 (s)
C-6	122.9	8.18 (d, $J = 8.10$ Hz)
C-7	120.4	7.26 (t, $J = 7.60$ Hz)
C-8	126.7	7.53 (t, $J = 7.60$ Hz)
C-9	110.0	7.68 (d, $J = 8.10$ Hz)
C-10	140.6	-
C-11	122.0	-
C-12	126.0	-
C-13	125.8	-
C-1'	134.2	-
C-2',C-6'	129.9	7.49 (m)
C-3',C-5'	128.6	7.46 (m)
C-4'	128.3	7.45 (m)

**<sup>1</sup>H and <sup>13</sup>C signal assignments of 4lg**



**COSY spectrum of 4lg**





HMBC spectrum of 4lg

<sup>1</sup> (a) N. Guimond, C. Gouliaras and K. Fagnou, *J. Am. Chem. Soc.* 2010, **132**, 6908; (b) S. Mochida, N. Umeda, K. Hirano, T. Satoh and M. Miura, *Chem. Lett.* 2010, **39**, 744; (c) G. Song, D. Chen, C.-L. Pan, R. H. Crabtree and X. Li, *J. Org. Chem.* 2010, **75**, 7487.

<sup>2</sup> Reddy, M. C.; Manikandan, R.; Jeganmohan, M. *Chem. Commun.* **2013**, 49, 6060

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