Electronic Supplementary Material (ESI) for Dalton Transactions.

Contrasting a Series of Bidentate Amido Phosphine Oxide, Sulfide, or Selenide Ligands and Complexes of Dimethyl Aluminum and Indium

Electronic Supporting Information

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1 - Synthesis of Precursors, Ligands, and Group 13 Complexes

Synthesis of Compound 1 - Dipp-N=C(CH_3)-(CH_2)P(C_6H_5)$_2$

Part 1: Following a modified previously established synthesis, a solution of 100 mL (0.5 mol) 2,6-diisopropylaniline in 200 mL (2.7 mol) acetone was refluxed for one week with ~40 mL anhydrous $MgSO_4$ to encourage condensation, producing N-isopropyl-N-2,6-diisopropylphenylimine with incomplete conversion possibly due to acid catalysed hydrolysis of the imine. The resulting solution was filtered through calcined diatomaceous earth, acetone removed, and the mixture was distilled *in vacuo* affording a 1:2 ratio of the amine to imine. Toluene, molecular sieves, and anhydrous calcium sulphate with refluxing over the course of a few weeks caused a 1:10 ratio of reagent to product. Distillation resulted in 3 fractions with the purest being the first, 9 grams of a 1:11 ratio of amine to imine.

Part 2: In a 500 mL round bottom flask 8.6 g was used directly in a -78 °C reaction in 230 mL pentane with first 0.5 g TMEDA (43 mmol) followed by addition of 17.7 mL of a 2.4 M n-BuLi solution (42 mmol) with slow warming to room temperature over 2 hours. Cooling back to -78 °C is followed by addition of 8.7 g chlorodiphenylphosphine (39 mmol) with warming to room temperature and stirring under nitrogen for two days. Filtration through calcined diatomaceous earth and removal of pentane *in vacuo* affords a yellowish white solid. Dissolution in minimal hot ethanol affords a golden solution which, upon cooling, crystallizes **1** as white crystal that can be isolated by filtration with 3×5 mL ethanol rinses. Concentration of the filtrate *in vacuo* can afford additional crops of crystals, and three were harvested with masses of 12.97 g, 0.77 g, and 0.05 g in order of collection. Purity decreased in later crops, with the first to third crop occurring at 91, 86, and 84% purity with an impurity of the oxide of **2**. The third crop presented as more suitable distinct single crystals for X-ray analysis compared to the first two crops that both occurred as highly radial spherical crystalline clumps.

Yield (if pure): Crop 1 = 82% Crop 2 = 5% Crop 3 = 0.32%

Yield (adjusted for oxide purity): Crop 1 = 75% Crop 2 = 4% Crop 3 = 0.27%

Synthesis of Compound 2 - Dipp-N=C(CH_3)-(CH_2)P(C_6H_5)$_2$O

In a 20 mL scintillation vial, a biphasic solution containing 5 mL toluene, 135 mg (0.34 mmol) **1**, and 1.5 mL 30% H_2O_2 (15 mmol) was mixed thoroughly for 3 hours. The layers were left to settle, and the organic layer was removed with a pipette. The aqueous layer was washed with 3×2 mL toluene and the organic fraction dried with $MgSO_4$. Filtering through calcined diatomaceous earth and evaporation *in vacuo* gave 140 mg (0.34 mmol, 99%) of an analytically pure white powder. Layering a toluene solution with pentane formed white radial needles, recrystallization from hot pentane gave colourless thin X-ray quality platy crystals, and acetone affords both plates and needles. The plate morphology from hot pentane form as the imine isomer, while the needles from acetone present as the enamine isomer.

Yield: 99%

m.p. (°C): 141.5 - 143.4 (Plate, imine).

Analytical Calc. for $C_{27}H_{32}NOP$: C: 77.67% H: 7.73% N: 3.35 P: 7.42% O: 3.83%.

Found: C: 77.49% H: 7.63% N: 3.22%.

Major isomer $CDCl_3$: imine, 84%:

^{1}H NMR ($CDCl_3$, 300 MHz, 25 °C): δ 7.87 – 7.94 (m, 4H, *m*-PPh_2), 7.46 – 7.58 (m, 6H, *o*-, *p*-PPh_2), 7.01 (m, 3H, *m*-Dipp, *p*-Dipp) 3.71 (d, 2H, $^2J_{P\text{-}H}$ = 14.2 Hz, PCH_2), 2.30 (sept, 2H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$), 1.92 (d, 3H, $^4J_{P\text{-}H}$ = 1.6 Hz, N=C(CH_3)), 0.93 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$), 0.92 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$) ppm.

$^{13}C\{^1H\}$ NMR ($CDCl_3$, 75 MHz, 25 °C): δ 164.93 (d, $^2J_{P\text{-}C}$ = 7.0 Hz, N=C), 145.39 (*ipso*-Dipp), 136.34 (*o*-Dipp), 132.89 (d, $^1J_{P\text{-}C}$ = 100.7 Hz, *ipso*-PPh_2), 131.95 (d, $^4J_{P\text{-}C}$ = 2.8 Hz, *p*-PPh_2), 130.94 (d, $^3J_{P\text{-}C}$ = 9.8 Hz, *m*-PPh_2), 128.73 (d, $^2J_{P\text{-}C}$ = 12.8 Hz, *o*-PPh_2), 123.65 (*p*-Dipp), 122.90 (*m*-Dipp), 44.16 (d, $^1J_{P\text{-}C}$ = 60.6 Hz, P*C*H_2), 27.73 (*C*H(CH_3)$_2$), 23.42 (CH(*C*H_3)$_2$), 23.25 (CH(*C*H_3)$_2$), 22.81 (s, N=C(*C*H_3)) ppm.
$^{31}P\{^1H\}$ NMR ($CDCl_3$, 121 MHz, 25 °C): δ 28.34 ppm.
Minor isomer $CDCl_3$: (E)-enamine, 11%:
^1H NMR ($CDCl_3$, 300 MHz, 25 °C): δ 8.80 (s, 1H, N-*H*), 7.76 - 7.83 (m, 4H, *m*-PPh_2), 4.27 (d, 1H, $^2J_{P\text{-}H}$ = 20.8 Hz, PC*H*), 3.17 (sept, 2H, $^3J_{H\text{-}H}$ = 6.8 Hz, C*H*(CH_3)$_2$), 1.63 (s, 3H, N=C(C*H*$_3$)), 1.18 (d, 6H, $^3J_{H\text{-}H}$ = 6.8 Hz, CH(C*H*$_3$)$_2$), 1.04 (d, 6H, $^3J_{H\text{-}H}$ = 6.8 Hz, CH(C*H*$_3$)$_2$) ppm.
$^{13}C\{^1H\}$ NMR ($CDCl_3$, 75 MHz, 25 °C): δ 162.37 (N=C), 147.56 (*ipso*-Dipp), 136.67 (d, $^1J_{P\text{-}C}$ = 103.5 Hz, *ipso*-PPh_2), 135.00 (*o*-Dipp), 130.86 (m, *m*-PPh_2), 128.38 (d, $^2J_{P\text{-}C}$ = 12.8 Hz, *o*-PPh_2), 123.86 (*p*-Dipp), 123.25 (*m*-Dipp), 76.26 (d, $^1J_{P\text{-}C}$ = 114.8 Hz, P*C*H), 28.29 (*C*H(CH_3)$_2$), 24.62 (CH(*C*H_3)$_2$), 21.81 (d, $^2J_{P\text{-}C}$ = 7.6 Hz, N=C(*C*H_3)) ppm.
$^{31}P\{^1H\}$ NMR ($CDCl_3$, 121 MHz, 25 °C): δ 29.72 ppm.
Minor isomer $CDCl_3$: (Z)-enamine, 5%:
^1H NMR ($CDCl_3$, 300 MHz, 25 °C): δ 5.27 (s, 1H, N-*H*), 4.04 (d, 1H, $^2J_{P\text{-}H}$ = 20.2 Hz, PC*H*), 3.17 (sept, 2H, $^3J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.27 (d, 6H, $^3J_{H\text{-}H}$ = 6.8 Hz, CH(C*H*$_3$)$_2$) ppm.
$^{31}P\{^1H\}$ NMR ($CDCl_3$, 121 MHz, 25 °C): δ 26.17 ppm.
Major isomer C_6D_6: imine, 80%:
^1H NMR (C_6D_6, 300 MHz, 25 °C): δ 7.82-7.89 (m, 4H, *m*-PPh_2), 7.03-7.13 (m, 9H, *o*-, *p*-PPh_2, *m*-Dipp, *p*-Dipp), 3.25 (d, 2H, $^2J_{P\text{-}H}$ = 14.5 Hz, PC*H*$_2$), 2.55 (sept, 2H, $^3J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.93 (d, 3H, $^4J_{P\text{-}H}$ = 1.6 Hz, N=C(C*H*$_3$)), 1.06 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(C*H*$_3$)$_2$), 1.02 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(C*H*$_3$)$_2$) ppm.
$^{13}C\{^1H\}$ NMR (C_6D_6, 75 MHz, 25 °C): δ 165.18 (d, $^2J_{P\text{-}C}$ = 7.7 Hz, N=C), 146.49 (*ipso*-Dipp), 136.53 (*o*-Dipp), 134.67 (d, $^1J_{P\text{-}C}$ = 99.5 Hz, *ipso*-PPh_2), 131.58 (d, $^4J_{P\text{-}C}$ = 2.7 Hz, *p*-PPh_2), 131.32 (d, $^3J_{P\text{-}C}$ = 9.6 Hz, *m*-PPh_2), 128.67 (d, $^2J_{P\text{-}C}$ = 11.6 Hz, *o*-PPh_2), 124.11 (*p*-Dipp), 123.37 (*m*-Dipp), 43.98 (d, $^1J_{P\text{-}C}$ = 61.8 Hz, P*C*H_2), 28.18 (*C*H(CH_3)$_2$), 23.74 (CH(*C*H_3)$_2$), 23.41 (CH(*C*H_3)$_2$), 21.72 (d, $^3J_{P\text{-}C}$ = 14.4 Hz, N=C(C*H*$_3$)) ppm.
$^{31}P\{^1H\}$ NMR (C_6D_6, 121 MHz, 25 °C): δ 25.38 ppm.
Minor isomer C_6D_6: (E)-enamine, 20%:
^1H NMR (C_6D_6, 300 MHz, 25 °C): δ 9.91 (s, 1H, N-*H*), 7.89 – 7.94 (m, m, 4H-PPh_2), 7.03-7.13 (m, 9H, *o*-, *p*-PPh_2, *m*-Dipp, *p*-Dipp), 4.27 (d, 1H, $^2J_{P\text{-}H}$ = 22.5 Hz, PC*H*), 3.36 (sept, 2H, $^3J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.56 (d, 3H, $^4J_{P\text{-}H}$ = 1.9 Hz, N=C(C*H*$_3$)), 1.13 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(C*H*$_3$)$_2$), 1.06 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(C*H*$_3$)$_2$) ppm.
$^{13}C\{^1H\}$ NMR (C_6D_6, 75 MHz, 25 °C): δ 162.47 (N=C), 147.93 (*o*-Dipp), 137.95 (d, $^1J_{P\text{-}C}$ = 102.7 Hz, *ipso*-Ph), 123.67 (*m*-Dipp), 77.22 (d, $^1J_{P\text{-}C}$ = 113.3 Hz, P*C*H), 28.80 (*C*H(CH_3)$_2$), 24.65 (CH(*C*H_3)$_2$), 22.67 (CH(*C*H_3)$_2$), 21.63 (N=C(*C*H_3)) ppm.
$^{31}P\{^1H\}$ NMR (C_6D_6, 121 MHz, 25 °C): δ 28.53 ppm.
Trace isomer C_6D_6: (Z)-enamine, <1%:
^1H NMR (C_6D_6, 300 MHz, 25 °C): δ 5.18 (bs, 1H, N-*H*)
$^{31}P\{^1H\}$ NMR (C_6D_6, 121 MHz, 25 °C): δ 22.26 ppm.

Major isomer CD_3CN: imine, 75%:
^1H NMR (C_6D_6, 300 MHz, 25 °C): δ 7.84-7.91 (m, 4H, *m*-PPh_2), 7.50-7.61 (m, 6H, *o*-, *p*-PPh_2), 6.93-7.04 (m, 3H, *m*-Dipp, *p*-Dipp) 3.75 (d, 2H, $^2J_{P\text{-}H}$ = 14.2 Hz, PC*H*$_2$), 2.35 (sept, 2H, $^3J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.80 (d, 3H, $^4J_{P\text{-}H}$ = 1.4 Hz, N=C(C*H*$_3$)), 0.96 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(C*H*$_3$)$_2$), 0.85 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(C*H*$_3$)$_2$) ppm.
$^{31}P\{^1H\}$ NMR (C_6D_6, 121 MHz, 25 °C): δ 26.82 ppm.
Minor isomer CD_3CN: (Z)-enamine 19%:

^{1}H NMR (CD_3CN, 300 MHz, 25 °C): δ 6.47 (s, 1H, N-*H*), 3.80 (m, 1H, PC*H*), 3.16 (sept, 2H, C*H*(CH_3)$_2$), 2.20 (s, 3H, N=C(C*H*$_3$)), 1.14-1.21 (12H, m, CH(C*H*$_3$)$_2$) ppm.
^{31}P{^{1}H} NMR (CD_3CN, 121 MHz, 25 °C): δ 28.61 ppm.
Minor isomer CD_3CN: (E)-enamine, 6%:
^{1}H NMR (CD_3CN, 300 MHz, 25 °C): δ 9.05 (s, 1H, N-*H*), 7.76 – 7.83 (m, m, 4H-PPh$_2$), 4.27 (d, 1H, $^{2}J_{P\text{-}H}$ = 20.8 Hz, PC*H*), 3.16 (sept, 2H, C*H*(CH_3)$_2$), 1.64 (s, 3H, N=C(C*H*$_3$)), 1.14-1.21 (m, 6H, CH(C*H*$_3$)$_2$), 1.02 (d, 6H, $^{3}J_{H\text{-}H}$ = 6.9 Hz, CH(C*H*$_3$)$_2$) ppm.
^{31}P{^{1}H} NMR (CD_3CN, 121 MHz, 25 °C): δ 22.74 ppm.

Synthesis of Compound 3 - Dipp-N=C(CH_3)CP(C_6H_5)$_2$S

To a 50 mL sealed reaction vessel under nitrogen, a solution containing ~10 mL toluene, 162 mg (0.40 mmol) **1** and 13 mg (0.40 mmol) S_8 were heated for 3.5 hr at 100 °C. Reduction to ~5 mL *in vacuo* and filtering through calcined diatomaceous earth gave a colourless solution. Evaporation of toluene *in vacuo* yields ~0.5 mL of a colourless oil turning to an analytically pure white crystalline solid after the addition of 10 mL pentane and removal *in vacuo* with gentle swirling and heating from a heat gun. Dissolution in minimal boiling pentane, slowly cooling to room temperature, cooling overnight at -18 °C followed by decanting and washing with minimal cold pentane twice and drying *in vacuo* produced X-ray quality thin rhombohedral prismatic colourless crystals. One large single rhombohedral monohydrate crystal was grown from slow evaporation of ethanol at room temperature open to air.
Yield: 97%.
m.p. (°C): 98.1 - 99.4.
Analytical Calc. for $C_{27}H_{32}NSP$: C: 74.79% H: 7.44% N: 3.23% P: 7.14% S: 7.40%. Found: C: 74.53%, H: 7.68%, N: 3.59%
Major isomer $CDCl_3$: imine, 75%:
^{1}H NMR ($CDCl_3$, 300 MHz, 25 °C): δ 8.00 – 8.05 (m, 4H, *m*-PPh$_2$), 7.43 – 7.53 (m, 6H, *o*-, *p*-PPh$_2$), 6.96 – 7.11 (m, 3H, *m*-Dipp, *p*-Dipp), 3.90 (d, 2H, $^{2}J_{P\text{-}H}$ = 15.0 Hz, PC*H*$_2$), 2.43 (sept, 2H, $^{3}J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.92 (d, 3H, $^{4}J_{P\text{-}H}$ = 1.6 Hz, N=C(C*H*$_3$)), 0.98 (d, 6H, $^{3}J_{H\text{-}H}$ = 6.9 Hz, CH(C*H*$_3$)$_2$), 0.92 (d, 6H, $^{3}J_{H\text{-}H}$ = 6.9 Hz, CH(C*H*$_3$)$_2$) ppm.
^{13}C{^{1}H} NMR ($CDCl_3$, 75 MHz, 25 °C): δ 164.56 (d, $^{2}J_{P\text{-}C}$ = 6.5 Hz, N=C), 145.60 (*ipso*-Dipp), 136.35 (*o*-Dipp), 132.94 (d, $^{1}J_{P\text{-}C}$ = 81.8 Hz, *ipso*-PPh$_2$), 131.70 (d, $^{4}J_{P\text{-}C}$ = 2.7 Hz, *p*-PPh$_2$), 131.53 (d, $^{3}J_{P\text{-}C}$ = 10.6 Hz, *m*-PPh$_2$), 128.68 (d, $^{2}J_{P\text{-}C}$ = 12.5 Hz, *o*-PPh$_2$), 123.69 (*p*-Dipp), 123.01 (*m*-Dipp), 46.16 (d, $^{1}J_{P\text{-}C}$ = 49.5 Hz, P*C*H$_2$), 27.83 (*C*H(CH_3)$_2$), 23.53 (CH(*C*H$_3$)$_2$), 23.37 (CH(*C*H$_3$)$_2$), 22.64 (N=C(C*H*$_3$)),) ppm.
^{31}P{^{1}H} NMR ($CDCl_3$, 121 MHz, 25 °C): δ 38.10 ppm.
Minor isomer $CDCl_3$: (E)-enamine, 19%:
^{1}H NMR ($CDCl_3$, 300 MHz, 25 °C): δ 8.55 (s, 1H, N-*H*), 7.90 – 7.97 (m, 4H, *m*-PPh$_2$), 4.43 (d, 1H, $^{2}J_{P\text{-}H}$ = 18.9 Hz, PC*H*), 3.09 (sept, 2H, $^{3}J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.70 (s, 3H, N=C(C*H*$_3$)), 1.19 (d, 6H, $^{3}J_{H\text{-}H}$ = 6.9 Hz, CH(C*H*$_3$)$_2$) ppm.
^{13}C{^{1}H} NMR ($CDCl_3$, 75 MHz, 25 °C): δ 160.81 (N=C), 147.3 (*ipso*-Dipp), 136.69 (*o*-Dipp), 135.10 (d, $^{1}J_{P\text{-}C}$ = 70.0 Hz), 130.98 (*p*-Ph), 124.05 (*p*-Dipp), 123.40 (*m*-Dipp), 76.29 (d, $^{1}J_{P\text{-}C}$ = 97.7 Hz, P*C*H), 28.47 (*C*H(CH_3)$_2$), 24.90 (CH(*C*H$_3$)$_2$), 23.73 (CH(*C*H$_3$)$_2$), 21.81 (d, $^{2}J_{P\text{-}C}$ = 14.4 Hz, N=C(*C*H$_3$)) ppm.
^{31}P{^{1}H} NMR ($CDCl_3$, 121 MHz, 25 °C): δ 27.96 ppm.
Minor isomer $CDCl_3$: (Z)-enamine, 6%:
^{1}H NMR ($CDCl_3$, 300 MHz, 25 °C): δ 5.25 (s, 1H, N-*H*), 3.72 (d, 1H, $^{2}J_{P\text{-}H}$ = 14.3 Hz, PC*H*), 2.31 (sept, 2H, $^{3}J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.62 (s, 3H, N=C(C*H*$_3$)), 0.91 – 1.00 (m, 12H, CH(C*H*$_3$)$_2$) ppm.
^{31}P{^{1}H} NMR ($CDCl_3$, 121 MHz, 25 °C): δ 28.34 ppm.

Trace isomer $CDCl_3$: ylide, <1%:
$^{31}P\{^{1}H\}$ NMR ($CDCl_3$, 121 MHz, 25 °C): δ 34.50 ppm.
Major isomer C_6D_6: imine, 84%:
^{1}H NMR (C_6D_6, 300 MHz, 25 °C): δ 7.93 – 8.01 (m, 4H, *m*-PPh_2), 6.98-7.12 (m, 9H, *o*-, *p*-PPh_2, *m*-Dipp, *p*-Dipp), 3.42 (d, 2H, $^{2}J_{P-H}$ = 14.9 Hz, PCH_2), 2.69 (sept, 2H, $^{3}J_{H-H}$ = 6.9 Hz, $CH(CH_3)_2$), 1.92 (d, 3H, $^{4}J_{P-H}$ = 1.8 Hz, N=C(CH_3)), 1.13 (d, 6H, $^{3}J_{H-H}$ = 6.9 Hz, $CH(CH_3)_2$), 1.01 (d, 6H, $^{3}J_{H-H}$ = 6.9 Hz, $CH(CH_3)_2$) ppm.
$^{31}P\{^{1}H\}$ NMR (C_6D_6, 121 MHz, 25 °C): δ 37.46 ppm.
Minor isomer C_6D_6: (E)-enamine, 16%:
^{1}H NMR (C_6D_6, 300 MHz, 25 °C): δ 9.37 (s, 1H, N-*H*), 8.01-8.09 (m, 4H, *m*-PPh_2), 6.98-7.12 (m, 9H, *o*-, *p*-PPh_2, *m*-Dipp, *p*-Dipp), 4.28 (d, 1H, $^{2}J_{P-H}$ = 18.5 Hz, PC*H*), 3.26 (sept, 2H, $^{3}J_{H-H}$ = 6.9 Hz, $CH(CH_3)_2$), 1.50 (s, 3H, N=C(CH_3)), 1.22 (d, 6H, $^{3}J_{H-H}$ = 6.9 Hz, $CH(CH_3)_2$) ppm.
$^{31}P\{^{1}H\}$ NMR (C_6D_6, 121 MHz, 25 °C): δ 28.13 ppm.
Major isomer CD_3CN: imine, 78%:
^{1}H NMR (CD_3CN, 300 MHz, 25 °C): δ 7.97 – 8.05 (m, 4H, *m*-PPh_2), 7.48 – 7.57 (m, 6H, *o*-, *p*-PPh_2), 6.94 – 7.05 (m, 3H, *m*-Dipp, *p*-Dipp), 3.99 (d, 2H, $^{2}J_{P-H}$ = 14.7 Hz, PCH_2), 2.51 (sept, 2H, $^{3}J_{H-H}$ = 6.9 Hz, $CH(CH_3)_2$), 1.83 (d, 3H, $^{4}J_{P-H}$ = 1.4 Hz, N=C(CH_3)), 1.01 (d, 6H, $^{3}J_{H-H}$ = 6.9 Hz, $CH(CH_3)_2$), 0.86 (d, 6H, $^{3}J_{H-H}$ = 6.9 Hz, $CH(CH_3)_2$) ppm.
$^{31}P\{^{1}H\}$ NMR (CD_3CN, 121 MHz, 25 °C): δ 38.10 ppm.
Minor isomer CD_3CN: (Z)-enamine, 10%:
^{1}H NMR (CD_3CN, 300 MHz, 25 °C): δ 6.49 (s, 1H, N-*H*), 3.80 (d, 1H, $^{2}J_{P-H}$ = 17.3 Hz, PC*H*), 3.07 (sept, 2H, $^{3}J_{H-H}$ = 6.8 Hz, $CH(CH_3)_2$), 1.80 (d, 3H, $^{3}J_{P-H}$ = 1.5 Hz, N=C(CH_3)), 1.23 (d, 6H, $^{3}J_{H-H}$ = 6.8 Hz, $CH(CH_3)_2$), 1.17 (d, 6H, $^{3}J_{H-H}$ = 6.8 Hz, $CH(CH_3)_2$) ppm.
$^{31}P\{^{1}H\}$ NMR (CD_3CN, 121 MHz, 25 °C): δ 27.39 ppm.
Minor isomer CD_3CN: (E)-enamine, 8%:
^{1}H NMR (CD_3CN, 300 MHz, 25 °C): δ 8.55 (s, 1H, N-*H*), 7.90 – 7.97 (m, 4H, *m*-PPh_2), 4.43 (d, 1H, $^{2}J_{P-H}$ = 18.9 Hz, PC*H*), 3.18 (sept, 2H, $^{3}J_{H-H}$ = 6.9 Hz, $CH(CH_3)_2$), 1.67 (s, 3H, N=C(CH_3)), 1.20 (d, 6H, $^{3}J_{H-H}$ = 6.9 Hz, $CH(CH_3)_2$), 0.93 (d, 6H, $^{3}J_{H-H}$ = 6.9 Hz, $CH(CH_3)_2$) ppm.
$^{31}P\{^{1}H\}$ NMR (CD_3CN, 121 MHz, 25 °C): δ 33.93 ppm.
Trace isomer CD_3CN: ylide, 4%:
^{1}H NMR (CD_3CN, 300 MHz, 25 °C): δ 3.75 (d, 1H, $^{2}J_{P-H}$ = 14.3 Hz, PC*H*), 2.36 (sept, 2H, $^{3}J_{H-H}$ = 6.8 Hz, $CH(CH_3)_2$) ppm.
$^{31}P\{^{1}H\}$ NMR (CD_3CN, 121 MHz, 25 °C): δ 26.91 ppm.

Synthesis of Compound 4 - Dipp-N=C(CH_3)CP(C_6H_5)$_2$Se

In a 50 mL sealed reaction vessel a solution containing ~20 mL toluene, 105 mg (0.26 mmol) **1** and 25 mg (0.32 mmol) grey selenium metal powder was prepared under nitrogen, sealed, and heated overnight at 100 °C. Filtering through calcined diatomaceous earth in open air and evaporation of toluene results in a yellowish white, finely crystalline pure powder with a 95% yield. Dissolution in minimal boiling pentane and cooling to -18 °C for 24hr produced radial tabular pale-yellow crystals affording 119 mg **4** (0.25 mmol). An unknown pink impurity was observed during some smaller scale reaction that may result from poor filtration of the fine selenium metal. A garlic odor attributed to selenium compounds was observed with prolonged storage under air.
Yield: 95%.
m.p. (°C): 81.8 - 83.4.
Analytical Calc. for $C_{27}H_{32}NSeP$: C: 67.49% H: 6.71% N: 2.92% P: 6.45% Se: 16.43%. Found: C: 67.53%, H: 6.74%, N: 2.87%

Major isomer $CDCl_3$: imine, 63%:
^{1}H NMR ($CDCl_3$, 300 MHz, 25 °C): δ 8.02 – 8.10 (m, 4H, *m*-PPh_2), 7.45 – 7.53 (m, 6H, *o*-, *p*-PPh_2), 7.00 – 7.10 (m, 3H, *m*-Dipp, *p*-Dipp), 4.10 (d, 2H, $^2J_{P\text{-}H}$ = 15.1 Hz, PCH_2), 2.51 (sept, 2H, $^3J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.98 (d, 3H, $^4J_{P\text{-}H}$ = 1.5 Hz, N=C(CH_3)), 1.04 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$), 0.96 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$) ppm.
^{13}C{^{1}H} NMR ($CDCl_3$, 75 MHz, 25 °C): δ 164.51 (d, $^2J_{P\text{-}C}$ = 6.7 Hz, N=C), 145.56 (ipso-Dipp), 136.28 (*o*-Dipp), 132.07 (d, $^3J_{P\text{-}C}$ = 10.7 Hz, *m*-PPh_2), 131.77 (d, $^4J_{P\text{-}C}$ = 2.8 Hz, *p*-PPh_2), 131.73 (d, $^1J_{P\text{-}C}$ = 73.0 Hz, *ipso*-PPh_2), 128.69 (d, $^2J_{P\text{-}C}$ = 12.4 Hz, *o*-PPh_2), 123.44 (*p*-Dipp), 123.03 (*m*-Dipp), 45.78 (d, $^1J_{P\text{-}C}$ = 42.8 Hz, P*C*H_2), 27.85 (*C*H(CH_3)$_2$), 23.97 (N=C(*C*H_3)), 23.58 (CH(*C*H_3)$_2$), 23.36 (CH(*C*H_3)$_2$) ppm.
^{31}P{^{1}H} NMR ($CDCl_3$, 121 MHz, 25 °C): δ 27.64 ($^1J_{Se\text{-}P}$ = 760 Hz) ppm.
^{77}Se NMR ($CDCl_3$, 57 MHz, 25 °C): δ -333 ($^1J_{Se\text{-}P}$ = 760 Hz) ppm.
Minor isomer $CDCl_3$: (E)-enamine, 33%:
^{1}H NMR ($CDCl_3$, 300 MHz, 25 °C): δ 8.43 (s, 1H, N-*H*), 7.97 – 8.01 (m, 4H, *m*-PPh_2), 7.11 – 7.23 (m, 3H, *m*-Dipp, *p*-Dipp), 4.51 (d, 1H, $^2J_{P\text{-}H}$ = 17.6 Hz, PC*H*), 3.14 (sept, 2H, $^3J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.76 (s, 3H, N=C(CH_3)), 1.24 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$), 1.01 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$) ppm.
^{13}C{^{1}H} NMR ($CDCl_3$, 75 MHz, 25 °C): δ 160.61 (N=*C*), 147.42 (*ipso*-Dipp), 134.70 (d, $^1J_{P\text{-}C}$ = 78.2 Hz, *ipso*-PPh_2), 134.52 (*o*-Dipp), 131.60 (d, $^3J_{P\text{-}C}$ = 11.3 Hz, *m*-PPh_2), 131.07 (d, $^4J_{P\text{-}C}$ = 2.9 Hz, *p*-PPh_2), 128.46 (d, $^2J_{P\text{-}C}$ = 12.7 Hz, *o*-PPh_2), 124.09 (*p*-Dipp), 123.71 (*m*-Dipp), 74.88 (d, $^1J_{P\text{-}C}$ = 89.9 Hz, P*C*H), 28.50 (*C*H(CH_3)$_2$), 25.10 (CH(*C*H_3)$_2$), 22.70 (CH(*C*H_3)$_2$), 22.42 (d, $^3J_{P\text{-}C}$ = 14.5 Hz, N=C(*C*H_3)) ppm.
^{31}P{^{1}H} NMR ($CDCl_3$, 121 MHz, 25 °C): δ 13.14 ($^1J_{Se\text{-}P}$ = 669 Hz) ppm.
^{77}Se NMR ($CDCl_3$, 57 MHz, 25 °C): δ -272 (d, $^1J_{Se\text{-}P}$ = 669 Hz) ppm.
Minor isomer $CDCl_3$: (Z)-enamine, 4%:
^{1}H NMR ($CDCl_3$, 300 MHz, 25 °C): δ 5.35 (bs, 1H, N-*H*), 3.76 (d, 1H, $^2J_{P\text{-}H}$ = 17.6 Hz, PC*H*), 3.20 (sept, 2H, $^3J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.67 (s, 3H, N=C(CH_3)), 1.31 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$) ppm.
^{31}P{^{1}H} NMR ($CDCl_3$, 121 MHz, 25 °C): δ 22.51 ppm.
Major isomer C_6D_6: imine, 74%:
^{1}H NMR (C_6D_6, 300 MHz, 25 °C): δ 7.95 – 8.02 (m, 4H, *m*-PPh_2), 6.97 – 7.13 (m, 9H, *o*-, *p*-PPh_2, *m*-Dipp, *p*-Dipp), 3.57 (d, 2H, $^2J_{P\text{-}H}$ = 15.1 Hz, PCH_2), 2.73 (sept, 2H, $^3J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.95 (d, 3H, $^4J_{P\text{-}H}$ = 1.7 Hz, N=C(CH_3)), 1.15 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$), 1.01 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$) ppm.
^{31}P{^{1}H} NMR (C_6D_6, 121 MHz, 25 °C): δ 27.13 ($^1J_{Se\text{-}P}$ = 760 Hz) ppm.
^{77}Se NMR (C_6D_6, 57 MHz, 25 °C): δ -333 ($^1J_{Se\text{-}P}$ = 760 Hz) ppm.
Minor isomer C_6D_6: (E)-enamine, 24%:
^{1}H NMR (C_6D_6, 300 MHz, 25 °C): δ 9.23 (s, 1H, N-*H*), 8.02 – 8.11 (m, 4H, *m*-PPh_2), 6.97 – 7.13 (m, 9H, *o*-, *p*-PPh_2, *m*-Dipp, *p*-Dipp), 4.39 (d, 1H, $^2J_{P\text{-}H}$ = 16.9 Hz, PC*H*), 3.26 (sept, 2H, $^3J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.50 (s, 3H, N=C(CH_3)), 1.13 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$), 1.01 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$) ppm.
^{31}P{^{1}H} NMR (C_6D_6, 121 MHz, 25 °C): δ 12.87 ($^1J_{Se\text{-}P}$ = 677 Hz) ppm.
^{77}Se NMR (C_6D_6, 57 MHz, 25 °C): δ -272 ($^1J_{Se\text{-}P}$ = 677 Hz) ppm.
Trace isomer C_6D_6: (Z)-enamine, 2%:
^{1}H NMR (C_6D_6, 300 MHz, 25 °C): δ 4.57 (bs, 1H, N-*H*), 3.79 (s, 1H, $^2J_{P\text{-}H}$ = 15.8 Hz, PC*H*), 3.10 (sept, 2H, $^3J_{H\text{-}H}$ = 6.8 Hz, C*H*(CH_3)$_2$), 2.08 (s, 3H, N=C(CH_3)), 1.36 (d, 6H, $^3J_{H\text{-}H}$ = 6.8 Hz, CH(CH_3)$_2$), 0.80 (d, 6H, $^3J_{H\text{-}H}$ = 6.8 Hz, CH(CH_3)$_2$) ppm.
^{31}P{^{1}H} NMR (C_6D_6, 121 MHz, 25 °C): δ 21.91 ppm.
Major isomer CD_3CN: imine, 72%:
^{1}H NMR (CD_3CN, 300 MHz, 25 °C): δ 7.97 – 8.04 (m, 4H, *m*-PPh_2), 7.48 – 7.56 (m, 6H, *o*-, *p*-PPh_2), 6.94 – 7.05 (m, 3H, *m*-Dipp, *p*-Dipp), 4.15 (d, 2H, $^2J_{P\text{-}H}$ = 15.0 Hz, PCH_2), 2.55 (sept, 2H, $^3J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.85 (d, 3H, $^4J_{P\text{-}H}$ = 1.2 Hz,

N=C(CH_3)), 1.02 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$), 0.86 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$) ppm.
^{31}P{^{1}H} NMR (CD_3CN, 121 MHz, 25 °C): δ 27.99 ($^1J_{Se\text{-}P}$ = 743 Hz) ppm.
Minor isomer CD_3CN: (E)-enamine, 14%:
^{1}H NMR (CD_3CN, 300 MHz, 25 °C): δ 8.63 (s, 1H, N-*H*), 7.73 – 7.79 (m, 6H, *o*-, *p*-PPh_2), 7.15 – 7.32 (m, 3H, *m*-Dipp, *p*-Dipp), 3.78 (d, 1H, $^2J_{P\text{-}H}$ = 17.2 Hz, PC*H*), 3.19 (sept, 2H, $^3J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 2.15 (s, 3H, N=C(CH_3)), 1.19 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$), 0.92 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$) ppm.
^{31}P{^{1}H} NMR (CD_3CN, 121 MHz, 25 °C): δ 12.56 ($^1J_{Se\text{-}P}$ = 661 Hz) ppm.
Minor isomer CD_3CN: (Z)-enamine, 13%:
^{1}H NMR (CD_3CN, 300 MHz, 25 °C): δ 7.89 – 7.97 (m, m, 4H-PPh_2), 7.48 – 7.56 (m, 6H, *o*-, *p*-PPh_2), 7.15 – 7.32 (m, 3H, *m*-Dipp, *p*-Dipp), 6.54 (s, 1H, N-*H*), 4.60 (d, 1H, $^2J_{P\text{-}H}$ = 17.8 Hz, PC*H*), 3.07 (sept, 2H, $^3J_{H\text{-}H}$ = 6.8 Hz, C*H*(CH_3)$_2$), 2.15 (s, 3H, N=C(CH_3)), 1.24 (d, 6H, $^3J_{H\text{-}H}$ = 6.8 Hz, CH(CH_3)$_2$), 1.17 (d, 6H, $^3J_{H\text{-}H}$ = 6.8 Hz, CH(CH_3)$_2$) ppm.
^{31}P{^{1}H} NMR (CD_3CN, 121 MHz, 25 °C): δ 22.42 ppm.
Trace isomer CD_3CN: ylide, 1%:
^{1}H NMR (CD_3CN, 300 MHz, 25 °C): δ 3.75 (d, 1H, $^2J_{P\text{-}H}$ = 17.8 Hz, PC*H*), 2.36 (sept, 2H, $^3J_{H\text{-}H}$ = 6.8 Hz, C*H*(CH_3)$_2$), 1.81 (s, 3H, N=C(CH_3)), 0.96 (d, 6H, $^3J_{H\text{-}H}$ = 6.8 Hz, CH(CH_3)$_2$), 0.65 (d, 6H, $^3J_{H\text{-}H}$ = 6.8 Hz, CH(CH_3)$_2$) ppm.

Synthesis of Compound 5 - Dipp-N(Al^-Me_2)=C(CH_3)CP^+(C_6H_5)$_2$O

In the glovebox within a 20 mL scintillation vial, 73 mg (0.17 mmol) **2** was dissolved in ~4 mL benzene, followed by addition of two aliquots of 0.45 mL for a total of 0.90 mL $AlMe_3$ (2.0 M, heptane, 1.80 mmol) using 2 insulin syringes, dispensing directly into the benzene solution. This was left to stir for 5 days, pumped mostly dry *in vacuo*, and transferred to an NMR tube using triplicate benzene-d_6 aliquots. The sample was put back into the dry box having shown complete PNMR conversion, transferred to a 20 mL vial and the solvent stripped *in vacuo*. The white residue with hints of beige was dissolved entirely into ~8 mL pentane with stirring and was then filtered into a pre-weighed vial through a calcined diatomaceous earth pipette filter with triplicate pentane washings totalling ~12 mL. Placing this in the freezer caused crystals to form that were re-dissolved into the bulk solution with gentle low heat from a heat gun. Once re dissolved, the solution was reduced to ~8 mL *in vacuo* with frequent observation until crystallization began. Redissolution with low heat and placing into the freezer similarly caused radial fine white needles to form. Decanting the bulk solution and rinsing the crystals with ~1 mL cold pentane and drying *in vacuo* gave 63 mg (0.13 mmol) when dry.
Yield: 76%.
m.p. (°C): 166.3 – 167.0.
Analytical Calc. for $C_{29}H_{37}NAlOP$: C: 73.55% H: 7.88% N: 2.96% Found: C: 73.55% H: 7.95% N: 3.08%.
^{1}H NMR (C_6D_6, 300 MHz, 25 °C): δ 7.63 – 7.70 (m, 4H, *m*-PPh_2), 6.98 – 7.16 (m, 9H, *m*-,*p*-Dipp, *o*-, *p*-PPh_2), 3.84 (d, 1H, $^2J_{P\text{-}H}$ = 26.3 Hz, PC*H*), 3.40 (sept, 2H, $^3J_{H\text{-}H}$ = 7.0 Hz, C*H*(CH_3)$_2$), 1.67 (d, 3H, $^4J_{P\text{-}H}$ = 1.3 Hz, N=C(CH_3)), 1.24 (d, 6H, $^3J_{H\text{-}H}$ = 7.0 Hz, CH(CH_3)$_2$), 1.05 (d, 6H, $^3J_{H\text{-}H}$ = 7.0 Hz, CH(CH_3)$_2$), -0.29 (6H, s, Al(CH_3)$_2$) ppm.
^{13}C{^{1}H} NMR (C_6D_6, 75MHz, 25 °C): δ 175.2 (s, N=*C*), 145.6 (s, *o*-Dipp), 142.6 (s, *ipso*-Dipp), 133.45 (d, $^1J_{P\text{-}C}$ = 111.0 Hz, *ipso*-PPh_2), 132.11 (d, $^4J_{P\text{-}C}$ = 2.4 Hz, *p*-PPh_2), 131.74 (d, $^3J_{P\text{-}C}$ = 10.6 Hz, *m*-PPh_2), 128.6 (d, $^2J_{P\text{-}C}$ = 12.5 Hz, *o*-PPh_2), 126.46 (s, *p*-Dipp), 124.04 (s, *m*-Dipp), 66.31 (d, $^1J_{P\text{-}C}$ = 103.3 Hz, P*C*H), 28.2 (s, *C*H(CH_3)$_2$), 25.67 (d, $^3J_{P\text{-}C}$ = 15.6 Hz, N=C(*C*H_3)), 25.01 (s, CH(*C*H_3)$_2$), 24.46 (s, CH(*C*H_3)$_2$), -9.38 (s, Al(*C*H_3)$_2$) ppm.

^{27}Al NMR (C_6D_6, 78 MHz, 25 °C): δ 67 ppm +/- 1600 Hz.
^{31}P{^{1}H} NMR (C_6D_6, 121 MHz, 25 °C): δ 41.45 ppm.

Synthesis of Compound 6 - Dipp-N(Al$^-$Me$_2$)-C(CH$_3$)=(CH)P$^+$(C$_6$H$_5$)$_2$S

In a 20 mL scintillation vial 66 mg (0.15 mmol) **3** was dissolved in 2 mL toluene and added to a 50 mL sealed reaction vessel with a PTFE stopper followed by addition of 0.087 mL AlMe$_3$ (2.0 M, heptane, 0.17 mmol) added to 2 mL toluene prior with 3×2 mL rinsing totalling 10 mL toluene. Heating to 100 °C for 1.5 hours was followed by cooling to room temperature, filtering through a calcined diatomaceous earth pipette filter in the glovebox with triplicate rinsing into a pre-weighed 20 mL scintillation vial. Drying *in vacuo* and rinsing the crushed white solid with 2×1 mL pentane gave 62 mg (0.13 mmol) of an analytically pure crystalline white solid. Colourless prismatic crystals were grown from cooling a pentane solution of **6**.
Yield: 83%
m.p. (°C): 149.9-151.3
Analytical Calc. for $C_{29}H_{37}NAlSP$: C: 71.14% H: 7.62% N: 2.85% Found: C: 71.22% H: 7.63% N: 2.92%
^{1}H NMR (C_6D_6, 300 MHz, 25 °C): δ 7.68 – 7.75 (m, 4H, *m*-PPh$_2$), 7.10 – 7.19 (m, 3H, *m*-,*p*-Dipp), 6.98 – 7.05 (m, 6H, *o*-,*p*-PPh$_2$), 3.92 (d, 1H, $^2J_{\text{P-H}}$ = 18.7 Hz, PC*H*), 3.67 (sept, 2H, $^3J_{\text{H-H}}$ = 6.9 Hz, C*H*(CH$_3$)$_2$), 1.72 (d, 3H, $^4J_{\text{P-H}}$ = 1.7 Hz, N=C(C*H*$_3$)), 1.37 (d, 6H, $^3J_{\text{H-H}}$ = 6.9 Hz, CH(C*H*$_3$)$_2$), 1.24 (d, 6H, $^3J_{\text{H-H}}$ = 6.9 Hz, CH(C*H*$_3$)$_2$), -0.48 (6H, s, Al(C*H*$_3$)$_2$) ppm.
^{13}C{^{1}H} NMR (C_6D_6, 75 MHz, 25 °C): δ 170.3 (s, N=C), 145.8 (*o*-Dipp), 142.8 (*ipso*-Dipp), 134.9 (d, $^1J_{\text{P-C}}$ = 90.2 Hz, *ipso*-PPh$_2$), 131.5 – 131.7 (m, *m*-,*p*-PPh$_2$), 128.6 (d, $^2J_{\text{P-C}}$ = 12.5 Hz, *o*-PPh$_2$), 126.6 (*p*-Dipp), 124.1 (*m*-Dipp), 66.1 (d, $^1J_{\text{P-C}}$ = 103.3 Hz, P*C*H), 28.2 (*C*H(CH$_3$)$_2$), 27.2 (d, $^3J_{\text{P-C}}$ = 15.6 Hz, N=C(*C*H$_3$)), 25.1 (CH(*C*H$_3$)$_2$), 24.6 (CH(*C*H$_3$)$_2$), -7.8 (Al(*C*H$_3$)$_2$) ppm.
^{27}Al NMR (C_6D_6, 78 MHz, 25 °C): δ 73 ppm +/- 2340 Hz.
^{31}P{^{1}H} NMR (C_6D_6, 121 MHz, 25 °C): δ 25.80 ppm.

Synthesis of Compound 7 - Dipp-N(Al$^-$Me$_2$)-C(CH$_3$)=(CH)P$^+$(C$_6$H$_5$)$_2$Se

In a 20 mL scintillation vial 33 mg (0.07 mmol) **4** was dissolved in 1 mL toluene followed by addition of 0.040 mL AlMe$_3$ (2.0 M, heptane, 0.08 mmol) below the liquid level to avoid decomposition. Triplicate rinsing followed mixing with an additional 5mL toluene into a 50 mL sealed reaction vessel with heating at 100 °C for 4 hours. The toluene was stripped *in vacuo* and the solids rinsed twice with 1 mL pentane. Dissolution in 2 mL toluene, filtering through a calcined diatomaceous earth pipette, and rinsing the flask and filter three times with 1 mL aliquots of toluene was followed by drying *in vacuo* giving 28 mg (0.05 mmol) of an analytically pure crystalline white solid. Colourless prismatic crystals were grown from cooling a pentane solution of **7**.
Yield: 77%
m.p. (°C): - 168.3-171.3.
Analytical Calc. for $C_{29}H_{37}NAlSeP$: C: 64.92% H: 6.95% N: 2.61% Found: C: 65.13% H: 6.95% N: 2.55%
^{1}H NMR (C_6D_6, 300 MHz, 25 °C): δ 7.69 – 7.77 (m, 4H, *m*-PPh$_2$, $^5J_{\text{Se-H}}$ = 80 Hz) , 7.10 – 7.19 (m, 3H, *p*-Dipp, *m*-Dipp), 6.98 – 7.03 (m, 6H, *o*-, *p*-PPh$_2$), 4.03 (d, 1H, $^2J_{\text{P-H}}$ = 16.2 Hz, PC*H*), 3.70 (sept, 2H, $^3J_{\text{H-H}}$ = 6.9 Hz, C*H*(CH$_3$)$_2$), 1.76 (d, 3H, $^4J_{\text{P-H}}$ = 1.8 Hz, N=C(C*H*$_3$)), 1.38 (d, 6H, $^3J_{\text{H-H}}$ = 6.9 Hz, CH(C*H*$_3$)$_2$), 1.25 (d, 6H, $^3J_{\text{H-H}}$ = 6.9 Hz, CH(C*H*$_3$)$_2$), -0.49 (6H, s, Al(C*H*$_3$)$_2$) ppm.

$^{13}C\{^1H\}$ NMR (C_6D_6, 75 MHz, 25 °C): δ 170.1 (d, $^3J_{P-C}$ = 2.2 Hz, N=C), 145.9 (*o*-Dipp), 142.9 (*ipso*-Dipp), 134.3 (d, $^1J_{P-C}$ = 83.0 Hz, *ipso*-PPh_2), 132.0 (d, $^3J_{P-C}$ = 11.2 Hz, *m*-PPh_2), 131.6 (d, $^4J_{P-C}$ = 3.1 Hz, *p*-PPh_2), 128.7 (d, $^2J_{P-C}$ = 12.8 Hz, *o*-PPh_2), 126.7 (*p*-Dipp), 124.1 (*m*-Dipp), 65.7 (d, $^1J_{P-C}$ = 96.6 Hz, P*C*H), 28.2 (*C*H($CH_3)_2$), 27.7 (d, $^3J_{P-C}$ = 15.8 Hz, N=C(*C*H_3)), 25.2 (CH(*C*$H_3)_2$), 24.6 (CH(*C*$H_3)_2$), -7.2 (Al(*C*$H_3)_2$) ppm.
^{27}Al NMR (C_6D_6, 78 MHz, 25 °C): δ 71 ppm +/- 1550 Hz.
$^{31}P\{^1H\}$ NMR (C_6D_6, 121 MHz, 25 °C): δ 10.14 ppm ($^1J_{Se-P}$ = 496 Hz).
^{77}Se NMR (C_6D_6, 57 MHz, 25 °C): δ -274.8 ppm ($^1J_{Se-P}$ = 496 Hz).

Synthesis of Compound 8 -Dipp-N(In^-Me_2)-C(CH_3)(CH)$P^+(C_6H_5)_2$O

Both 0.2250 g (0.54 mmol) **3** and 0.0881 g (0.55 mmol) $InMe_3$ were transferred to a 50 mL sealed reaction vessel along with 10 mL toluene. The reaction mixture was heated for 3 hours at 110 °C, cooled to room temperature and transferred into a glovebox. The solution was concentrated *in vacuo* to 5 mL, filtered through a pipette filter (3×2 mL toluene rinses) into a pre-weighed 20 mL scintillation vial and concentrated to a thick oil. Addition and removal *in vacuo* of 3×1 mL pentane to the oil resulted in a white solid forming that was dissolved in minimal pentane (4 mL) with heat in the sealed flask, followed by cooling to -35 °C overnight. The following day small white radial crystals had formed, and the solution was allowed to slowly evaporate to 2 mL and warm to room temperature causing growth of pink-hued larger bladed crystals growing from the central white core determined to be a 6-membered chelate by SC-XRD. The solids were rinsed once with 1 mL pentane and dried *in vacuo* totaling 0.2592 g. ^1H NMR analysis of one clump of crystals after drying showed 1.1 equivalents of pentane remained within the radial crystal, so prior to elemental analysis the sample was crushed. Assuming the whole sample to have the same pentane ratio of 1.1 eq. within the crystals, the adjusted yield is 0.227 g (0.40 mmol) of **8**. A much more dilute solution in pentane cooled to -35 °C formed small colourless blocks that formed as dimers.
Yield: 75%
m.p. (°C): 90.1 - 92.3
Analytical Calc. for: $C_{29}H_{37}NInOP$: 62.04% H: 6.64% N: 2.49% Found: C: 62.43% H: 6.77% N: 2.37%
^1H NMR (C_6D_6, 300 MHz, 25 °C): δ 7.77 – 7.84 (m, 4H, *m*-PPh_2), 7.02-7.11 (m, 9H, *m*-Dipp, *p*-Dipp, *o*-, *p*-PPh_2), 3.84 (d, 2H, $^2J_{P-H}$ = 24.1 Hz, PC*H*), 3.23 (sept, 2H, $^3J_{H-H}$ = 6.9 Hz, C*H*($CH_3)_2$), 1.71 (d, 3H, $^4J_{P-H}$ = 2.1 Hz,), 1.03-1.07 (12H, m, CH(C*H*$_3)_2$), 0.11 (6H, s, In(C*H*$_3)_2$) ppm.
$^{13}C\{^1H\}$ NMR (C_6D_6, 75 MHz, 25 °C): δ 175.3 (N=C), 144.7 (*ipso*-Dipp), 144.5 (*o*-Dipp), 136.4 (d, $^1J_{P-C}$ = 109.2 Hz, *ipso*-PPh_2), 132.0 (d, $^3J_{P-C}$ = 11.3 Hz, *m*-PPh_2), 131.4 (d, $^4J_{P-C}$ = 2.9 Hz, *p*-PPh_2), 128.5 (d, $^2J_{P-C}$ = 12.6 Hz, *o*-PPh_2), 125.6 (*p*-Dipp), 123.8 (*m*-Dipp), 66.9 (d, $^1J_{P-C}$ = 123.4 Hz, P*C*H), 28.0 (*C*H($CH_3)_2$), 27.4 (d, N=C(*C*H_3)), 25.2 (CH(*C*$H_3)_2$), 24.5 (CH(*C*$H_3)_2$), -6.7 (In(*C*$H_3)_2$ ppm.
$^{31}P\{^1H\}$ NMR (C_6D_6, 121 MHz, 25 °C): δ 36.54 ppm.

Synthesis of Compound 9 - Dipp-N(In^-Me_2)-C(CH_3)(CH)$P^+(C_6H_5)_2$S

Both 0.1842 g (0.42 mmol) **3** and 0.0694 g (0.43 mmol) $InMe_3$ were combined in a 50 mL sealed reaction vessel along with 10 mL toluene with subsequent heating to 100 °C for 5.5 hours. The mixture was left to stand for 4 days, and ^{31}P NMR analysis of the bulk solution with a few drops of C_6D_6 showed incomplete conversion of **3** to **9** and the mixture appeared to have a small amount of fine white precipitate, assumed to be $InMe_3$ hydrolysis products. To the reaction mixture an additional 0.0018 g (1 μmol) $InMe_3$ was added along with the NMR sample and 3 mL toluene to rinse,

followed by ~10 minutes of heating with a heat gun until bubbling occurred. The toluene was then removed *in vacuo* resulting in a thick oil that solidified into a white solid upon addition and *in vacuo* removal of 2×1 mL pentane. 3 mL pentane was then added to the flask with the solid and sealed, fully dissolving with mild heat until boiling occurred. The solution was transferred to a pre-weighed 20 mL scintillation vial with 3×1 mL pentane extraction through a lint free wipe plug in a pipette. The vial was heated and sealed once boiling began and then placed in a -35 °C freezer over night. The bulk solution was removed with a pipette, 1 mL pentane used to rinse, and subsequent drying *in vacuo* gave 0.1830 mg of large radial white crystals. ^{1}H NMR analysis of one radial clump of crystals after drying showed 1.3 equivalents of pentane remained within the crystal clump, so prior to elemental analysis the sample was crushed. Assuming the whole sample to have the same pentane ratio of 1.3 eq. within the crystals, the adjusted yield is 0.157 g (0.27 mmol) of **9**.
Yield: 72%
m.p. (°C): 120.6-121.7
Analytical Calc. for $C_{29}H_{37}NInSP$: C: 60.32% H: 6.46% N: 2.43% Found: C: 60.43% H: 6.50% N: 2.37%.
^{1}H NMR (C_6D_6, 300 MHz, 25 °C): δ 7.79 – 7.86 (m, 4H, *m*-PPh_2), 7.10 (m, 3H, *m*-Dipp, *p*-Dipp), 7.03-7.06 (m, 6H, *o*-, *p*-PPh_2), 3.70 (d, 2H, $^2J_{P\text{-}H}$ = 18.8 Hz, PC*H*), 3.56 (sept, 2H, $^3J_{H\text{-}H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.78 (d, 3H, $^4J_{P\text{-}H}$ = 1.9 Hz, N=C(CH_3)), 1.27 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$), 1.22 (d, 6H, $^3J_{H\text{-}H}$ = 6.9 Hz, CH(CH_3)$_2$), -0.22 (6H, s, In(CH_3)$_2$) ppm.
^{13}C{^{1}H} (C_6D_6, 75 MHz, 25 °C): δ 171.02 (N=C), 145.09 (*ipso*-Dipp), 144.47 (*o*-Dipp), 136.80 (d, $^1J_{P\text{-}C}$ = 89.1 Hz, *ipso*-PPh_2), 131.63 (d, $^3J_{P\text{-}C}$ = 11.1 Hz, *m*-PPh_2), 136.80 (d, $^4J_{P\text{-}C}$ = 2.8 Hz, *p*-PPh_2), 128.56 (d, $^2J_{P\text{-}C}$ = 12.9 Hz, *o*-PPh_2), 125.70 (*p*-Dipp), 123.94 (*m*-Dipp), 62.70 (d, $^1J_{P\text{-}C}$ = 107.0 Hz, P*C*H), 34.45 (*C*H(CH_3)$_2$), 27.98 (CH(*C*H_3)$_2$), 26.97 (d, $^3J_{P\text{-}C}$ = 15.1 Hz, N=C(*C*H_3)), 25.10 (CH(*C*H_3)$_2$), -6.67 (In(*C*H_3)$_2$ ppm.
^{31}P{^{1}H} (C_6D_6, 121 MHz, 25 °C): δ 26.27 ppm.
^{1}H NMR (C_7D_8, 300 MHz, 25 °C): δ 7.75 – 7.83 (m, m, 4H-PPh_2), 6.97 – 7.09 (m, m, 9H-,*p*-Dipp, *o*-, *p*-PPh_2), 3.66 (d, 1H, $^2J_{P\text{-}H}$ = 18.8 Hz, PC*H*), 3.50 (sept, 2H, $^3J_{H\text{-}H}$ = 7.0 Hz, C(CH_3)$_2$*H*), 1.75 (d, 3H, $^4J_{P\text{-}H}$ = 2.0 Hz, N=C(CH_3)), 1.24 (d, 6H, $^3J_{H\text{-}H}$ = 7.0 Hz, C(CH_3)$_2$H), 1.20 (d, 6H, $^3J_{H\text{-}H}$ = 7.0 Hz, C(CH_3)$_2$H), -0.29 (s, 6H, In(CH_3)$_2$). ppm.

Synthesis of Compound 10 - Dipp-N(In^-Me_2)-C(CH_3)(CH)P^+(C_6H_5)$_2$Se

Both 0.2050 g (0.43 mmol) **4** and 0.0712 g (0.45 mmol) $InMe_3$ were added to a 50 mL sealed reaction vessel using 10 mL toluene. The solution was heated to 110 °C for 1.5 hr then cooled and left overnight. Heating the next day for an additional 2 hours was followed by cooling, transferring to the glovebox, and removing the toluene and methane *in vacuo* resulting in a thick gold oil. Addition and subsequent *in vacuo* removal of 3×1 mL of pentane gave a white solid precipitate and golden oil. 2 mL pentane was used to wash the solid, which crystallized upon slow evaporation. The remaining white solid was transferred to a 20 mL scintillation vial with pentane (5x2 mL), sealed, and warmed until the solid had completely dissolved. The vial was vented, heated until bubbling occurred, and sealed again and placed in -35 °C for 4 days. Decanting the pentane into the previous washings, rinsing with 1 mL cold pentane, and drying for 30 minutes prior to drying *in vacuo* left 0.1948 g slightly yellow radial prismatic crystals. ^{1}H NMR analysis of one radial clump of crystals after drying showed 0.06 equivalents of pentane remained within the crystal clump, so prior to elemental analysis the sample was crushed. Assuming the whole sample to have the same pentane ratio of 0.06 within the crystals, the adjusted yield is 0.193 g (0.31 mmol) of **10**.

Yield: 73%
m.p. (°C): 141.7-143.0
Analytical Calc. for $C_{29}H_{37}NInSeP$: C: 55.79% H: 5.97% N: 2.24% Found: C: 55.40% H: 6.12% N: 2.15%.
^{1}H (C_6D_6, 300 MHz, 25 °C): δ 7.79 – 7.87 (m, 4H, *m*-PPh_2), 7.11 (s, m, 3H-Dipp, *p*-Dipp), 7.01 – 7.05 (m, 6H, *o*-, *p*-PPh_2), 3.76 (d, 2H, $^{2}J_{P-H}$ = 17.4 Hz, PC*H*), 3.61 (sept, 2H, $^{3}J_{H-H}$ = 6.9 Hz, C*H*(CH_3)$_2$), 1.82 (d, 3H, $^{4}J_{P-H}$ = 2.2 Hz, N=C(CH_3)), 1.31 (d, 6H, $^{3}J_{H-H}$ = 6.9 Hz, CH(CH_3)$_2$), 1.25 (d, 6H, $^{3}J_{H-H}$ = 6.9 Hz, CH(CH_3)$_2$), -0.27 (6H, s, In(CH_3)$_2$) ppm.
^{13}C{^{1}H} (C_6D_6, 75 MHz, 25 °C): δ 170.7 (d, $^{2}J_{P-C}$ = 2.3 Hz, N=C), 145.3 (*ipso*-Dipp), 144.4 (*o*-Dipp), 136.0 (d, $^{1}J_{P-C}$ = 82.3 Hz, *ipso*-PPh_2), 132.0 (d, $^{3}J_{P-C}$ = 11.3 Hz, *m*-PPh_2), 131.4 (d, $^{4}J_{P-C}$ = 2.9 Hz, *p*-PPh_2), 128.6 (d, $^{2}J_{P-C}$ = 12.6 Hz, *o*-PPh_2), 125.7 (*p*-Dipp), 124.0 (*m*-Dipp), 61.0 (d, $^{1}J_{P-C}$ = 100.0 Hz, P*C*H), 28.0 (*C*H(CH_3)$_2$), 27.4 (d, $^{3}J_{P-C}$ = 15.8 Hz, N=C(*C*H_3)), 25.2 (CH(*C*H_3)$_2$), 24.5 (CH(*C*H_3)$_2$), -6.5 (In(*C*H_3)$_2$ ppm.
^{31}P{^{1}H} (C_6D_6, 121 MHz, 25 °C): δ 9.01 ($^{1}J_{Se-P}$ = 502 Hz) ppm.
^{77}Se (C_6D_6, 57 MHz, 25 °C): δ -293.5 ($^{1}J_{Se-P}$ = 502 Hz) ppm.

2 - NMR Spectroscopy

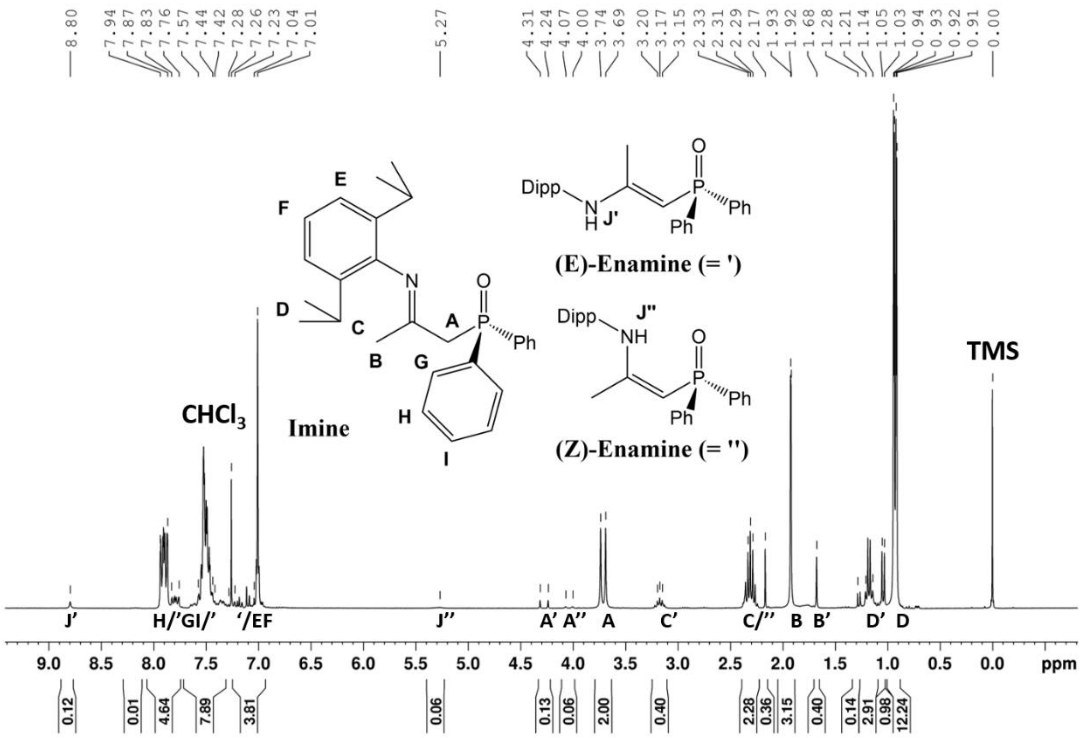


Figure S1 ^{1}H NMR spectrum of compound **2** recorded at 298K in $CDCl_3$.

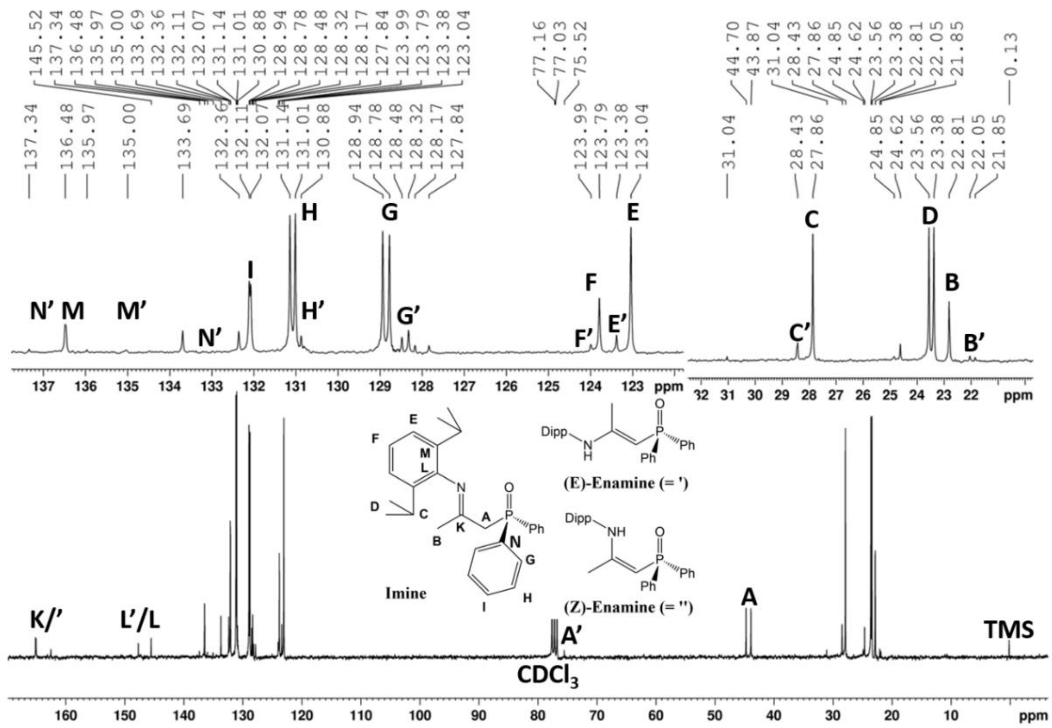


Figure S2 ^{13}C NMR spectrum of compound **2** recorded at 298K in $CDCl_3$

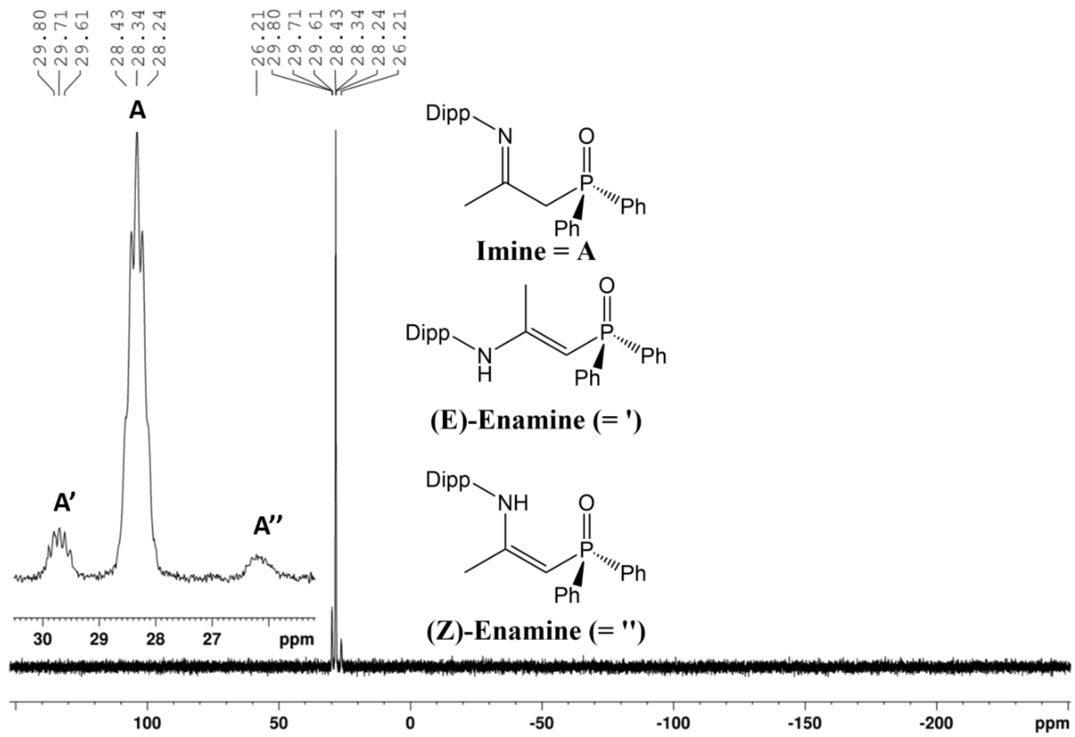


Figure S3 ^{31}P NMR spectrum (121 MHz) of compound **2** recorded at 298K in $CDCl_3$.

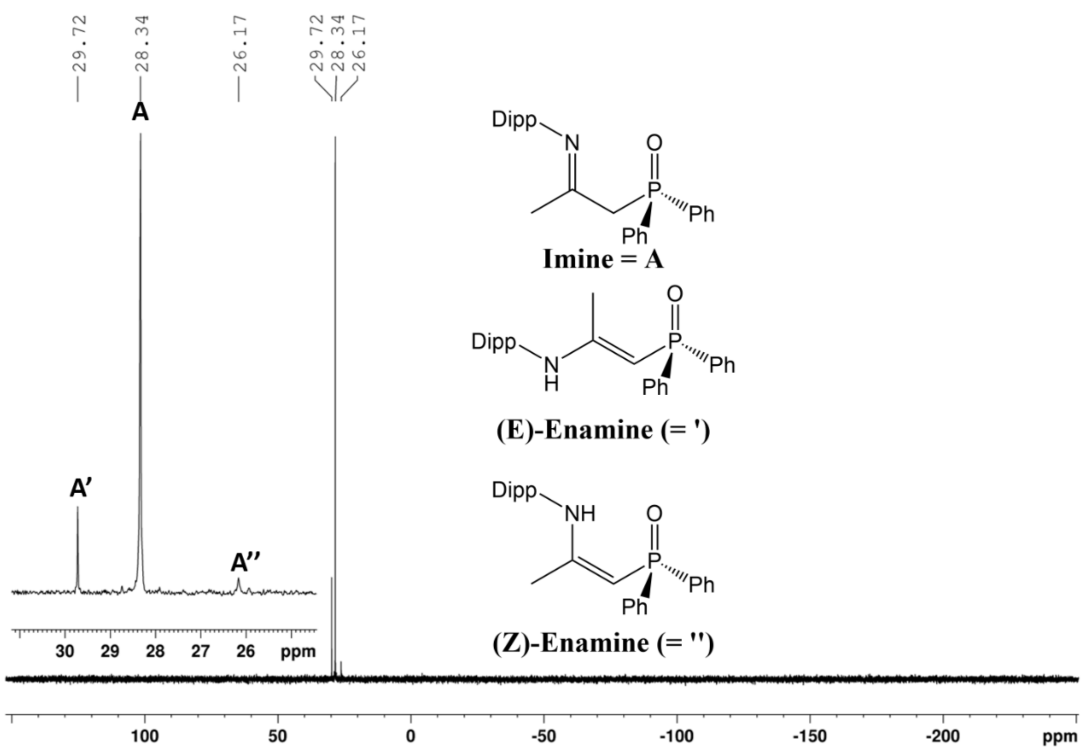


Figure S4 $^{31}P\{^1H\}$ NMR spectrum (121 MHz) of compound **2** recorded at 298K in $CDCl_3$.

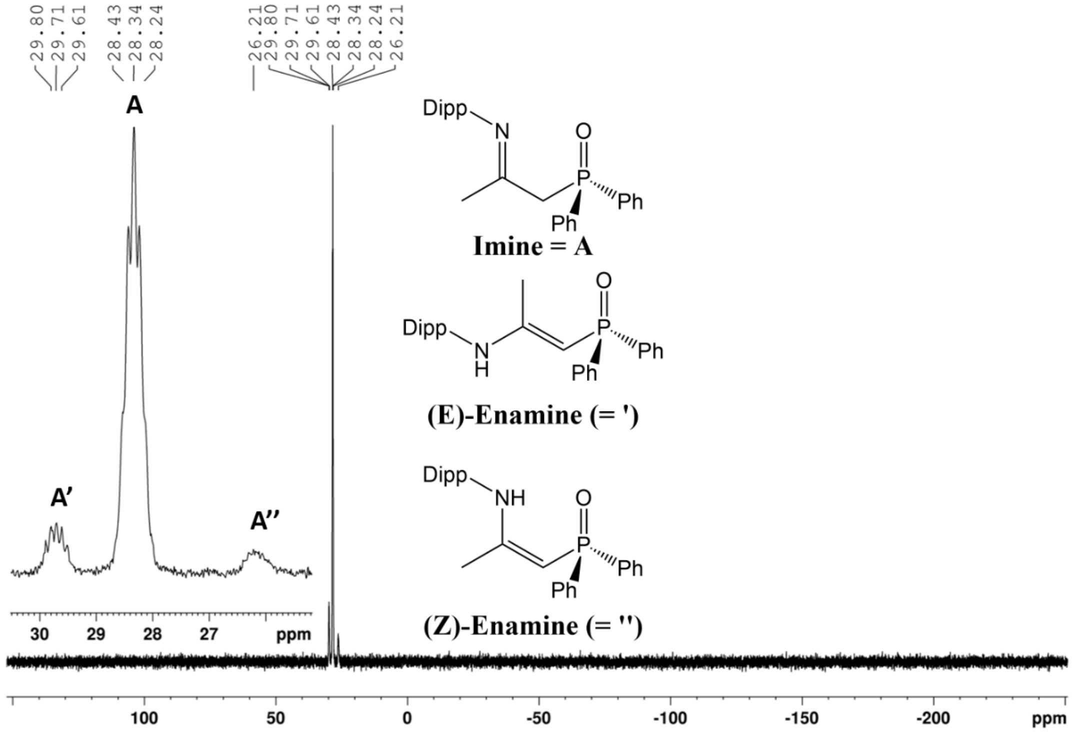


Figure S5 ^{31}P NMR spectrum (121 MHz) of compound **2** recorded at 298K in $CDCl_3$.

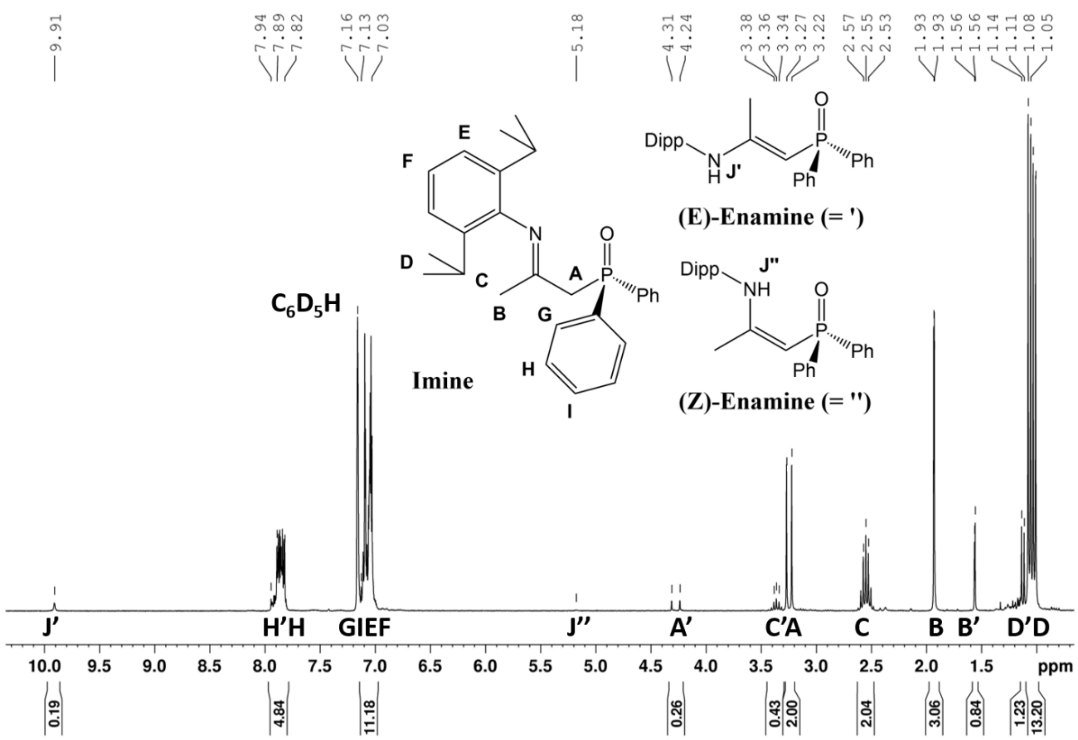


Figure S6 ^{1}H NMR spectrum of compound **2** recorded at 298K in C_6D_6.

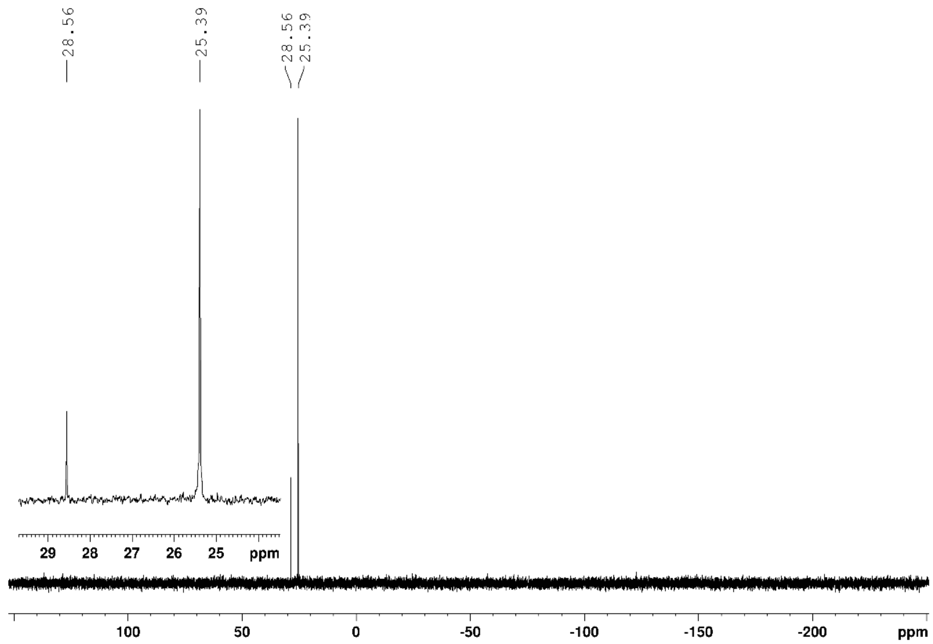


Figure S7 $^{31}P\{^{1}H\}$ NMR spectrum (121 MHz) of compound **2** recorded at 298K in C_6D_6.

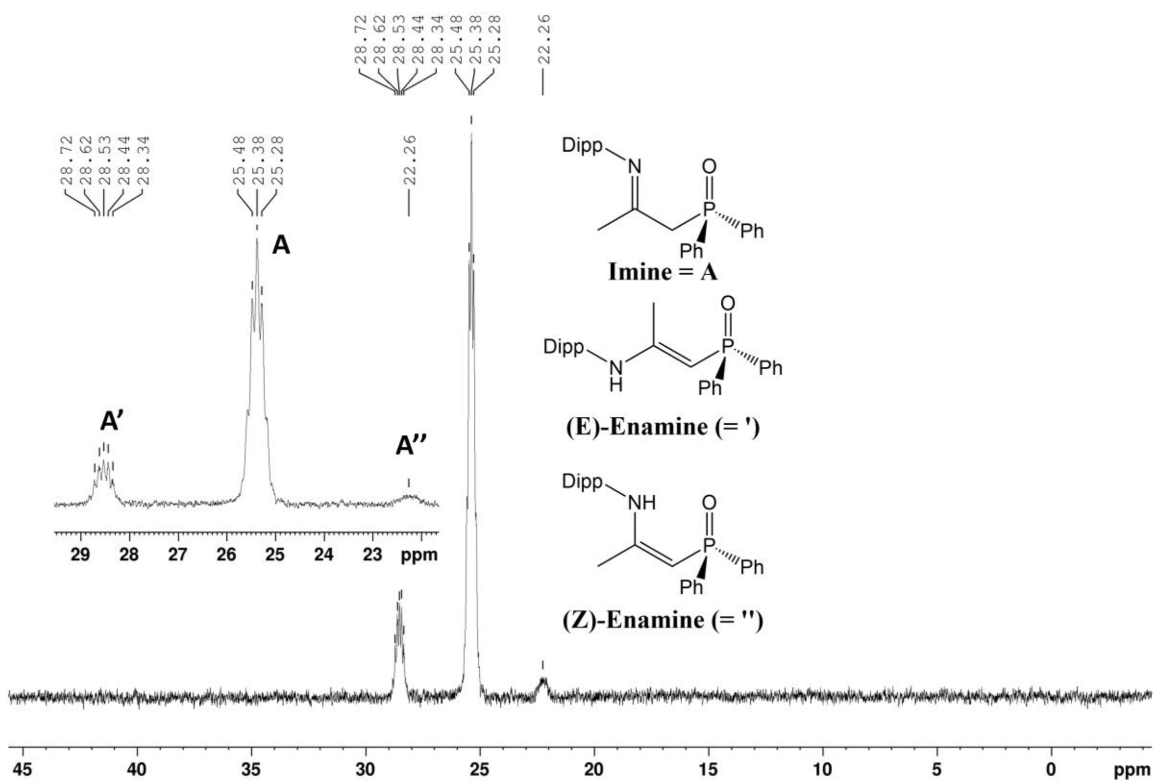


Figure S8 ^{31}P NMR spectrum (121 MHz) of compound **2** recorded at 298K in C_6D_6. Spectrum was acquired 6 hours after the $^{31}P\{^{1}H\}$ spectrum.

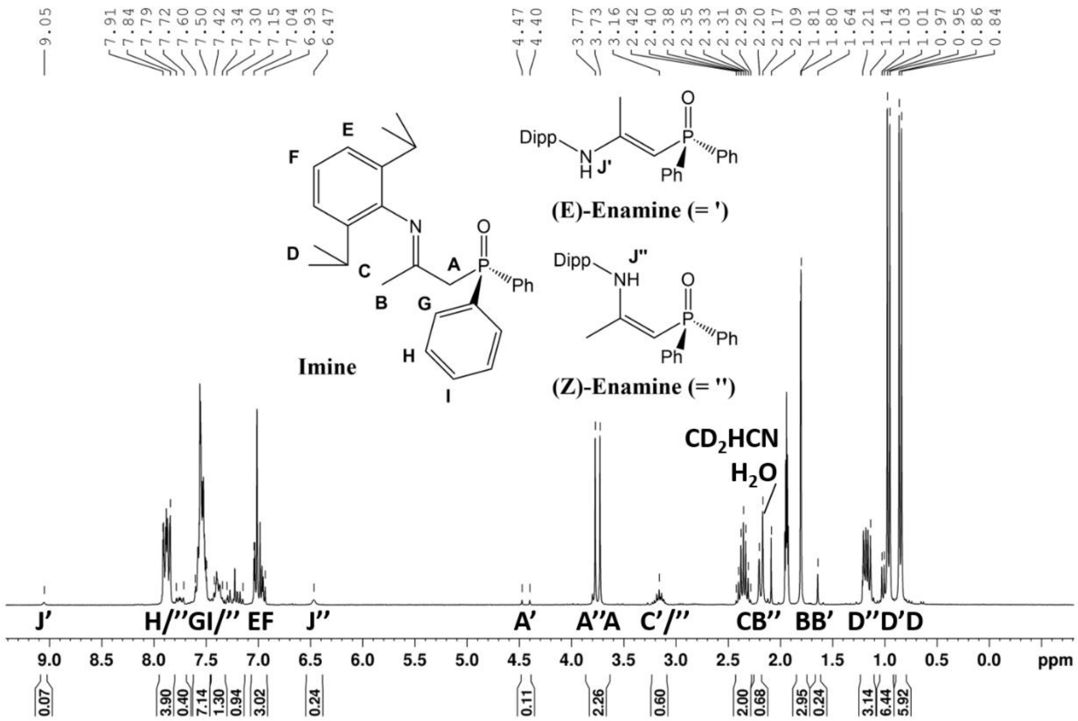


Figure S9 ^{1}H NMR spectrum of compound **2** recorded at 298K in CD_3CN.

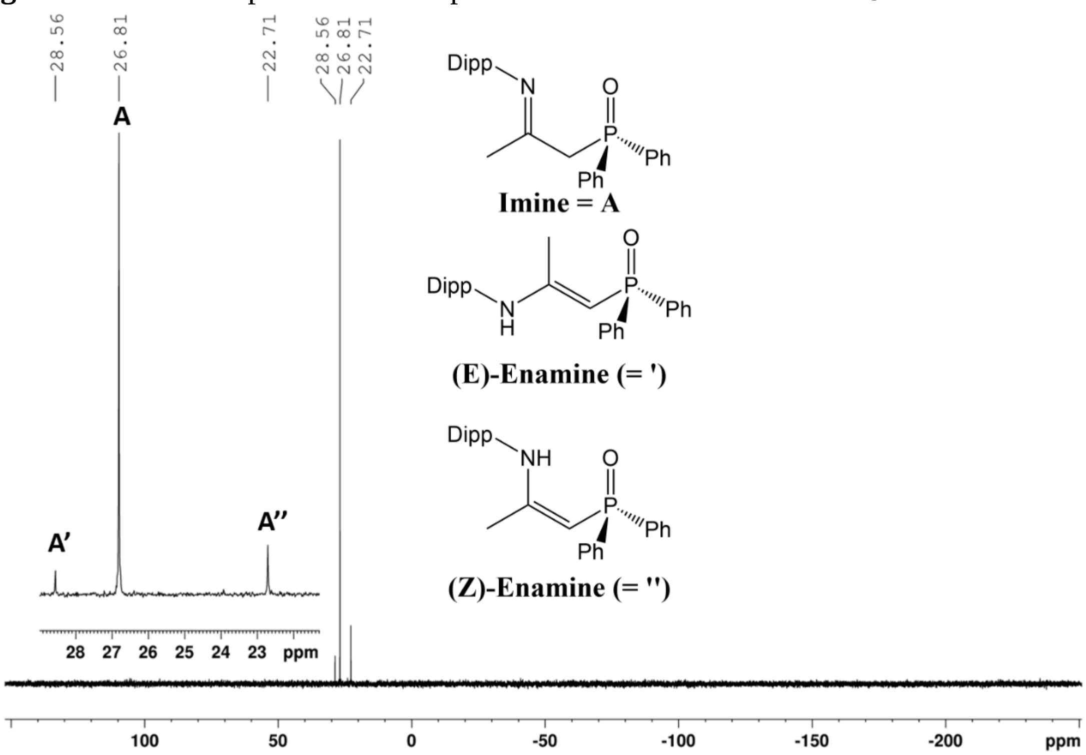


Figure S10 $^{31}P\{^{1}H\}$ NMR spectrum (121 MHz) of compound **2** recorded at 298K in CD_3CN.

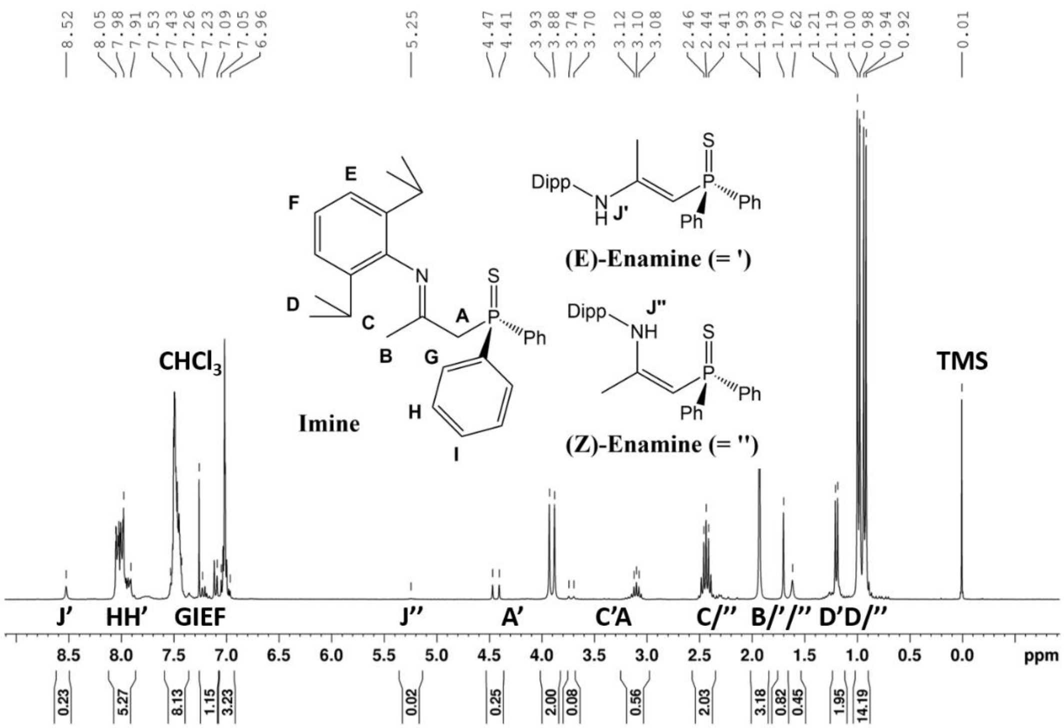


Figure S11 ^{1}H NMR spectrum of compound **3** recorded at 298K in $CDCl_3$.

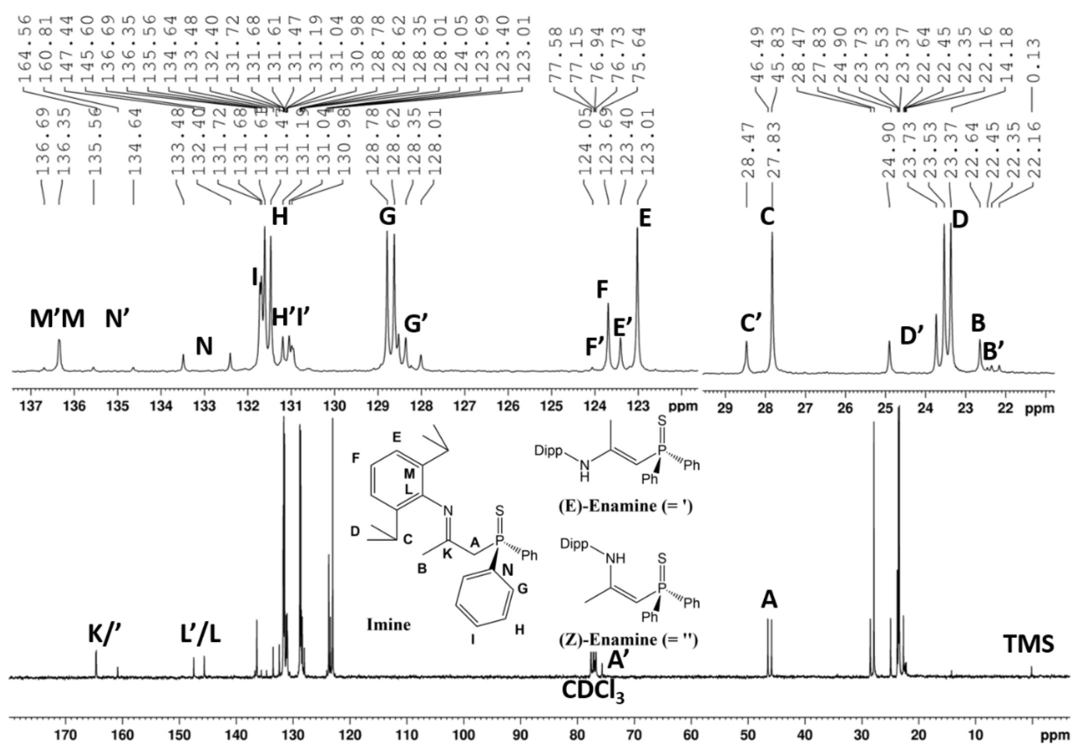


Figure S12 ^{13}C NMR spectrum of compound **3** recorded at 298K in $CDCl_3$.

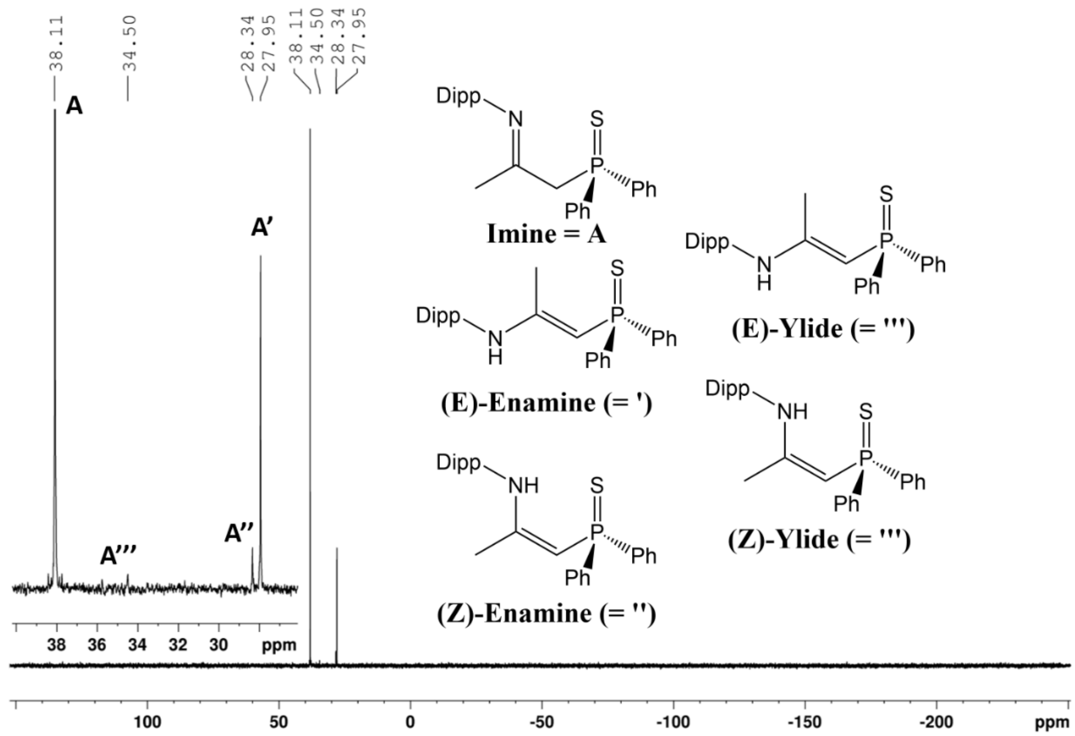


Figure S13 $^{31}P\{^{1}H\}$ NMR spectrum (121 MHz) of compound **3** recorded at 298K in $CDCl_3$.

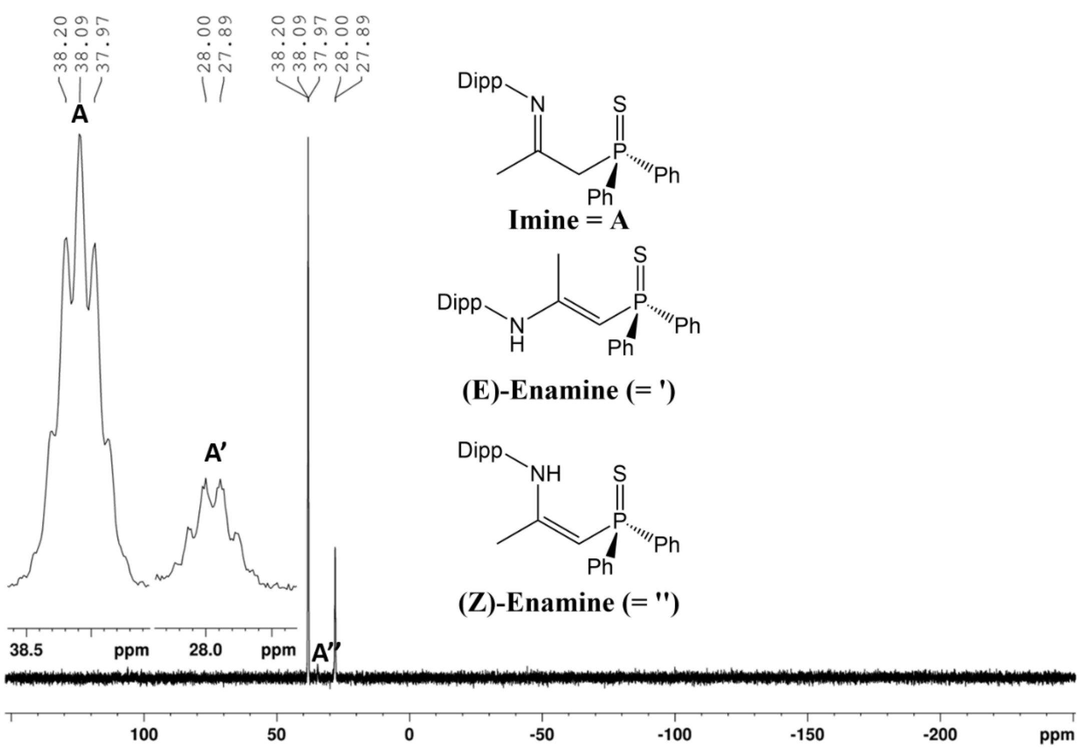


Figure S14 ^{31}P NMR spectrum (121 MHz) of compound **3** recorded at 298K in $CDCl_3$.

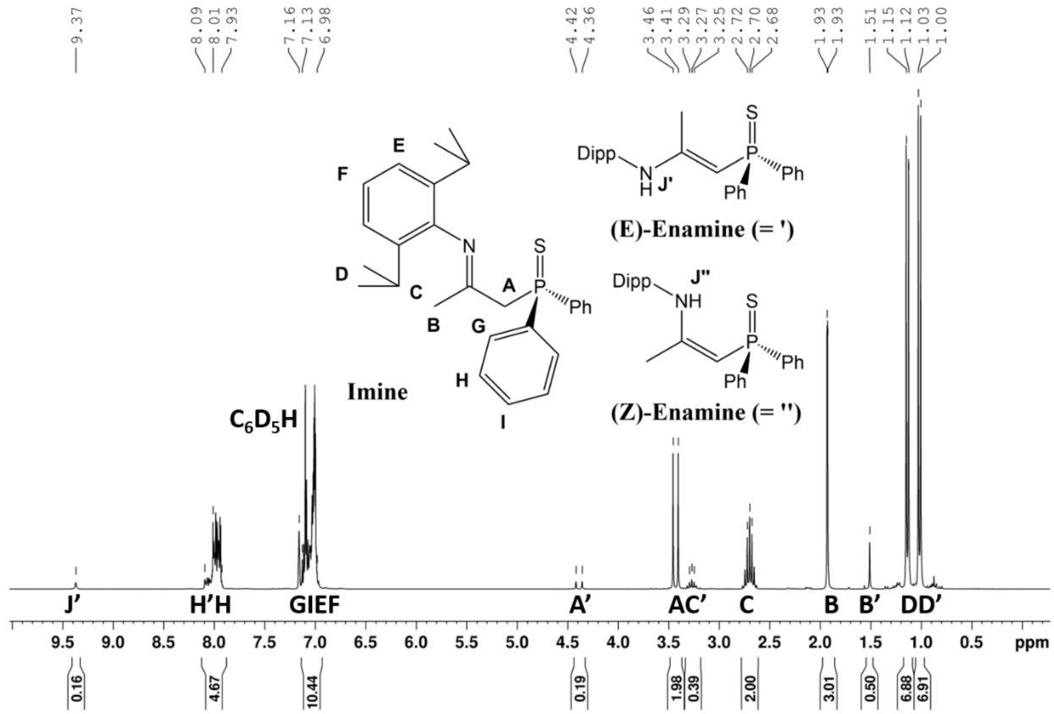


Figure S15 [1]H NMR spectrum of compound **3** recorded at 298K in C_6D_6.

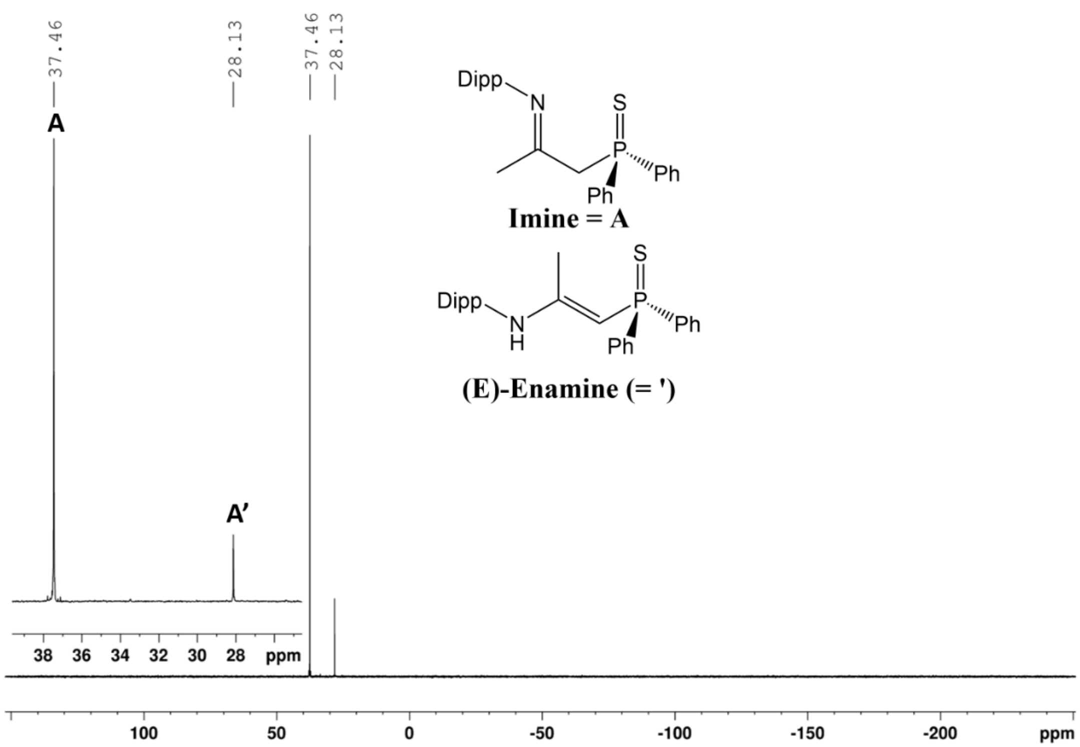


Figure S16 $^{31}P\{^{1}H\}$ NMR spectrum (121 MHz) of compound **3** recorded at 298K in C_6D_6.

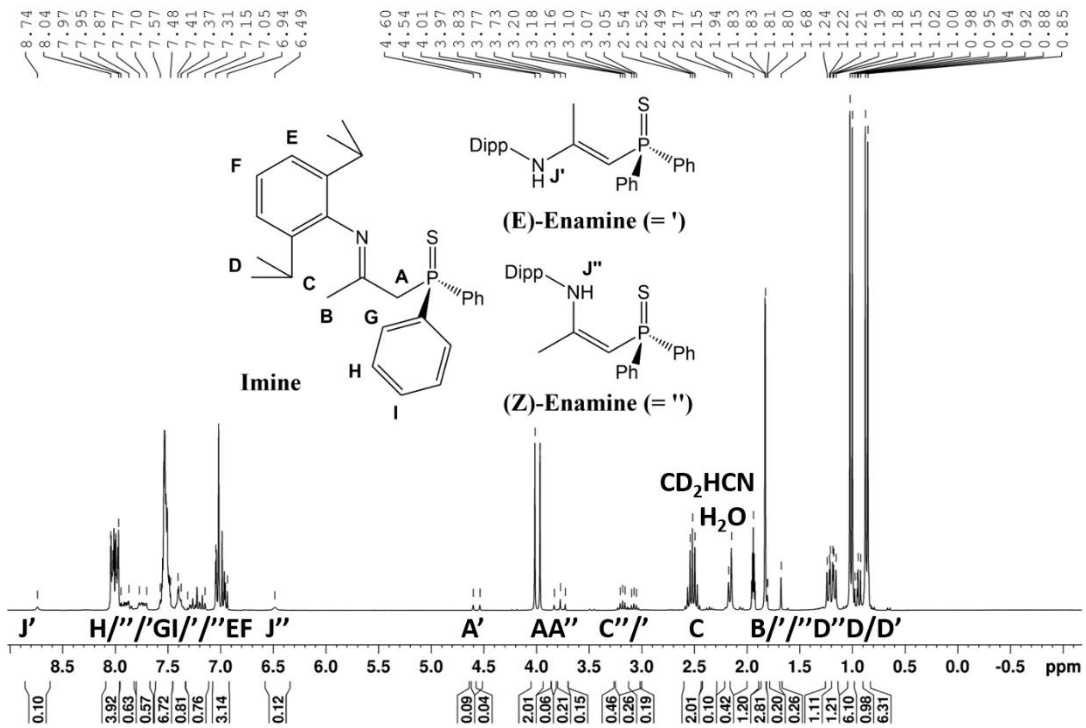


Figure S17 [1]H NMR spectrum of compound **3** recorded at 298K in CD_3CN.

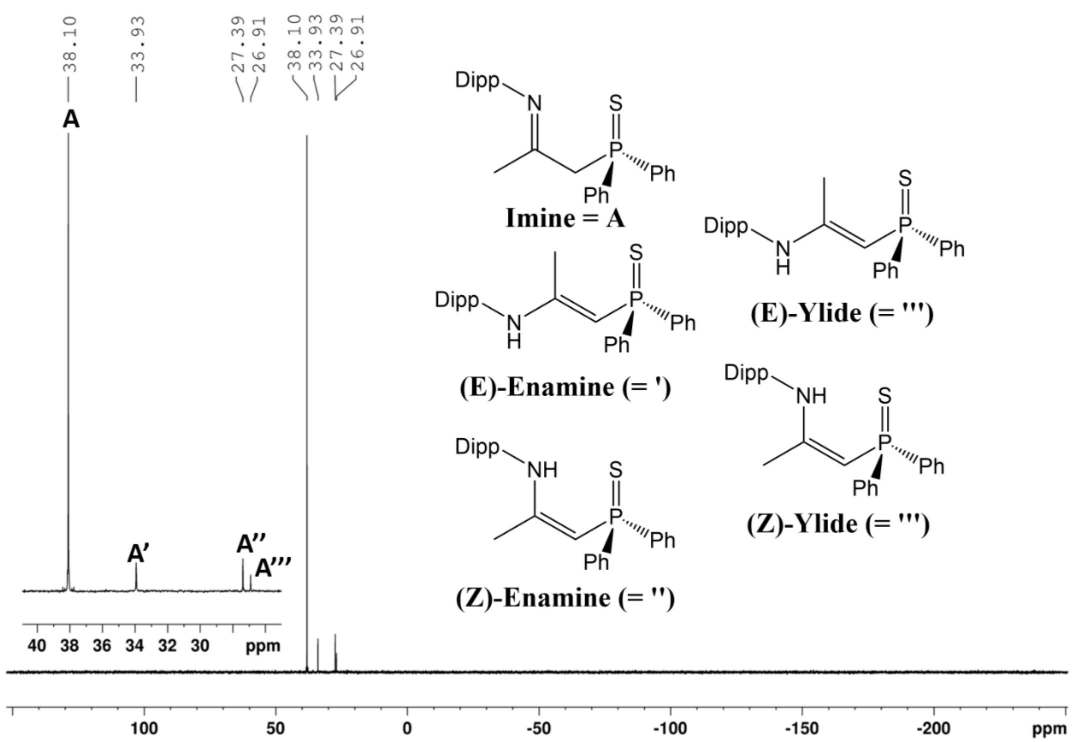


Figure S18 $^{31}P\{^1H\}$ NMR spectrum (121 MHz) of compound **3** recorded at 298K in CD_3CN.

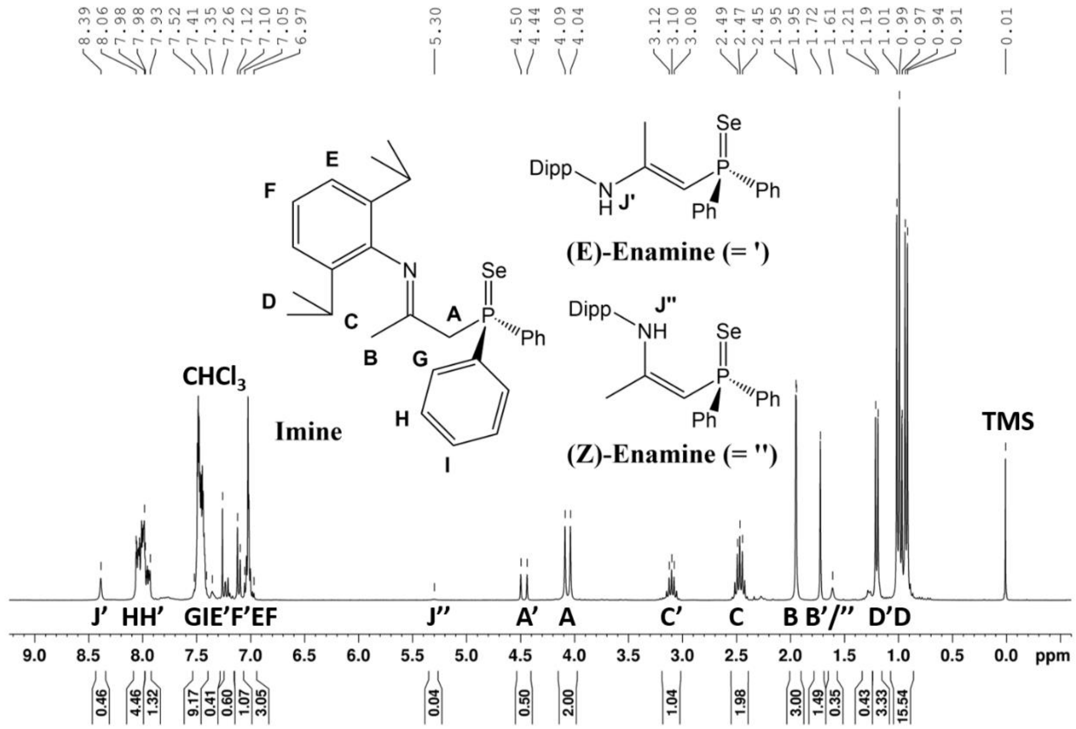


Figure S19 ^{1}H NMR spectrum of compound **4** recorded at 298K in $CDCl_3$.

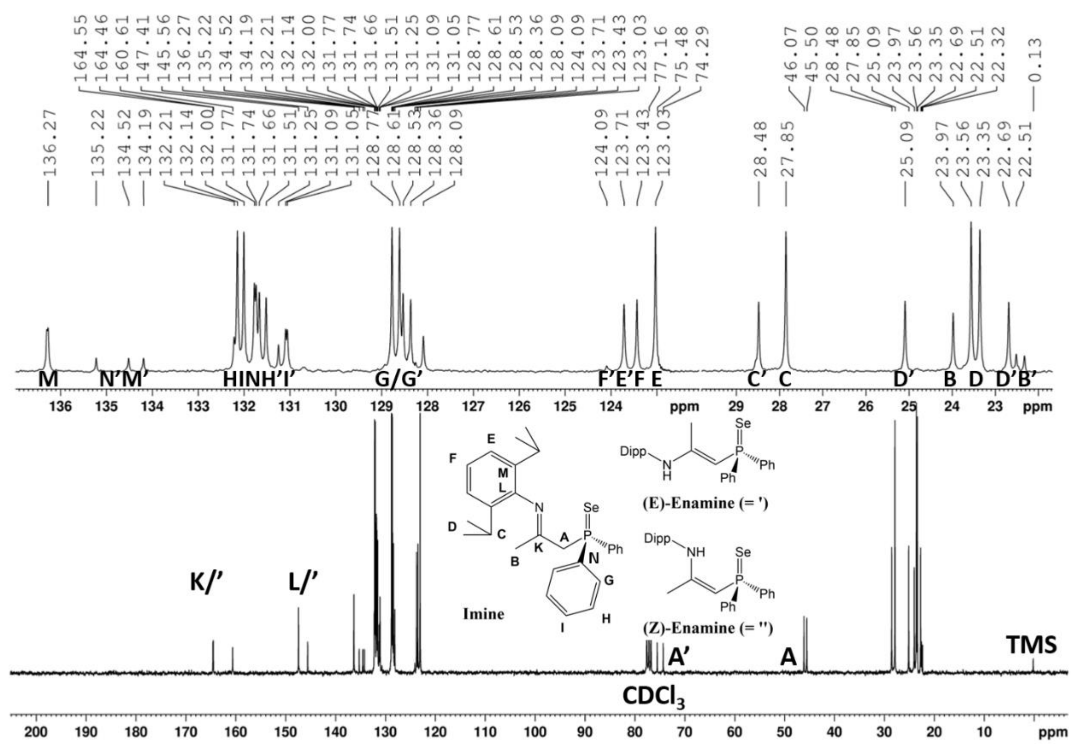


Figure S20 ^{13}C NMR spectrum of compound **4** recorded at 298K in $CDCl_3$.

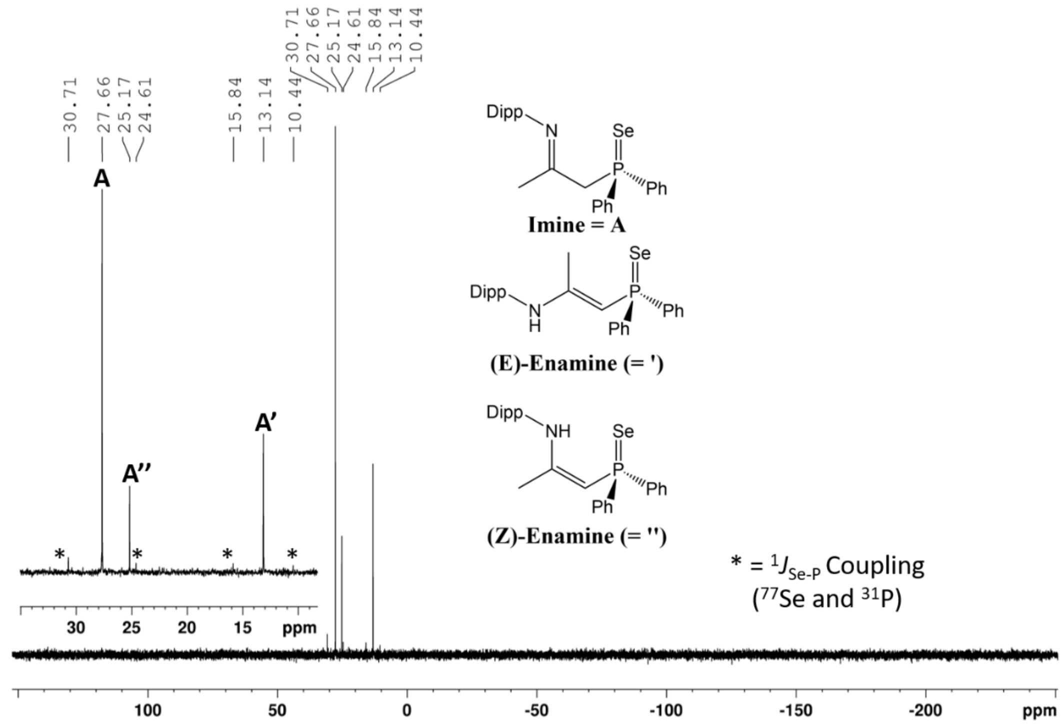


Figure S21 $^{31}P\{^{1}H\}$ NMR spectrum (121 MHz) of compound **4** recorded at 298K in $CDCl_3$.

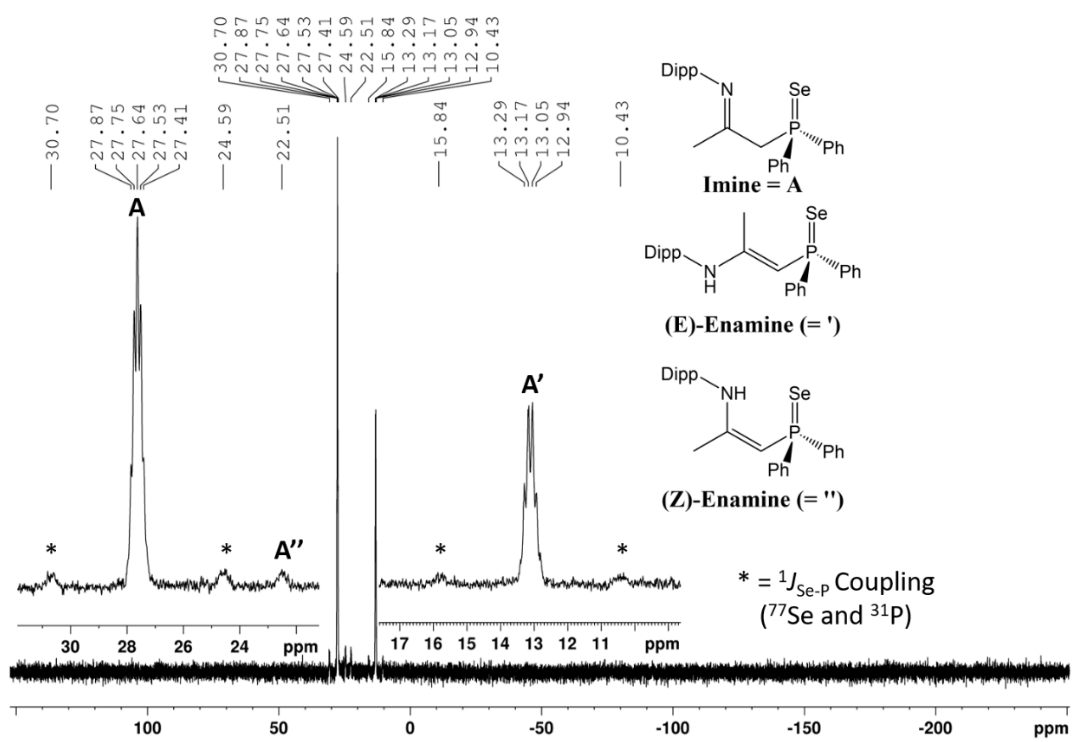


Figure S22 ^{31}P NMR spectrum (121 MHz) of compound **4** recorded at 298K in $CDCl_3$.

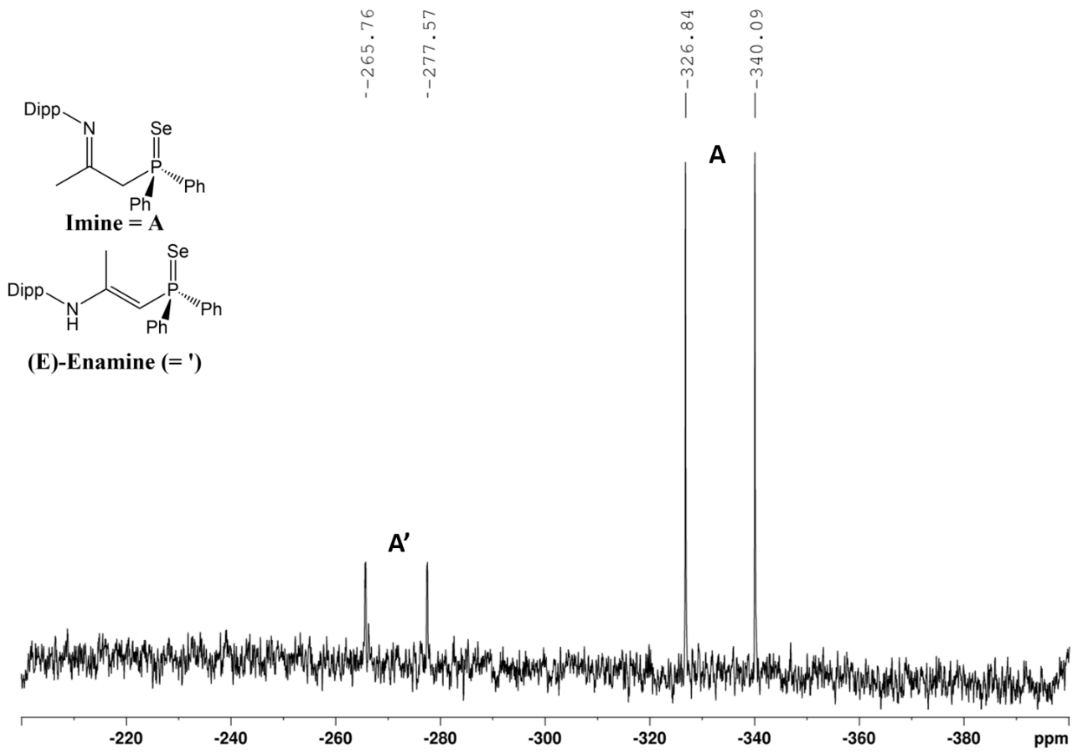


Figure S23 [77]Se NMR spectrum (57 MHz) of compound **4** recorded at 298K in $CDCl_3$.

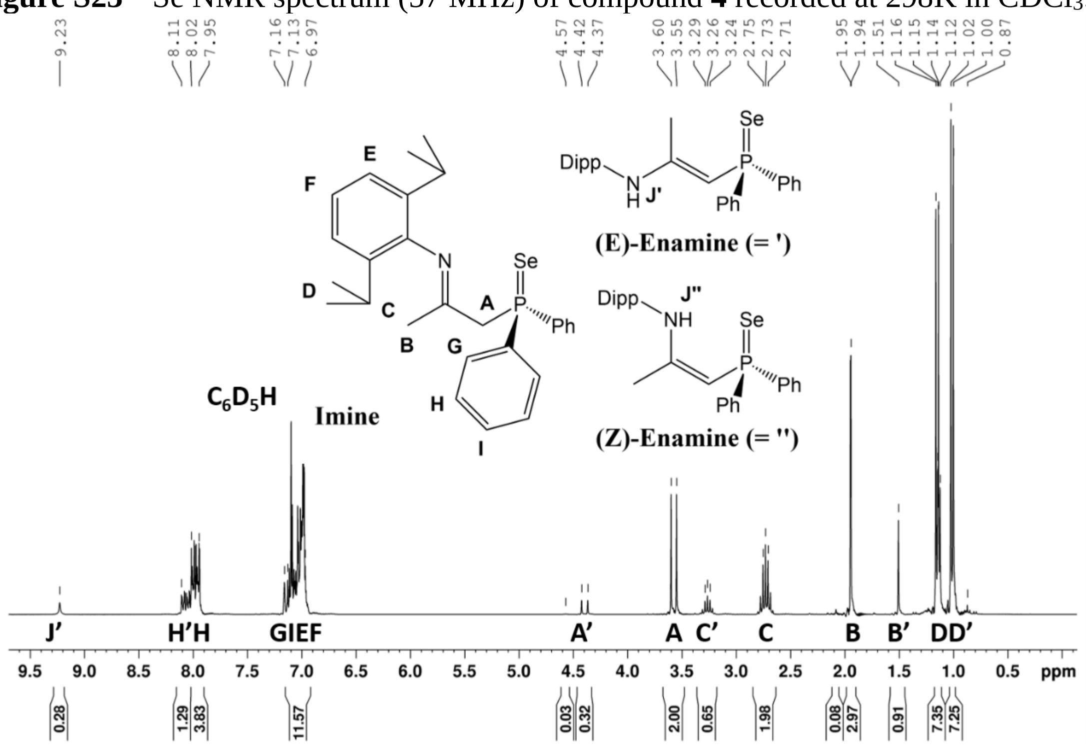


Figure S24 [1]H NMR spectrum of compound **4** recorded at 298K in C_6D_6.

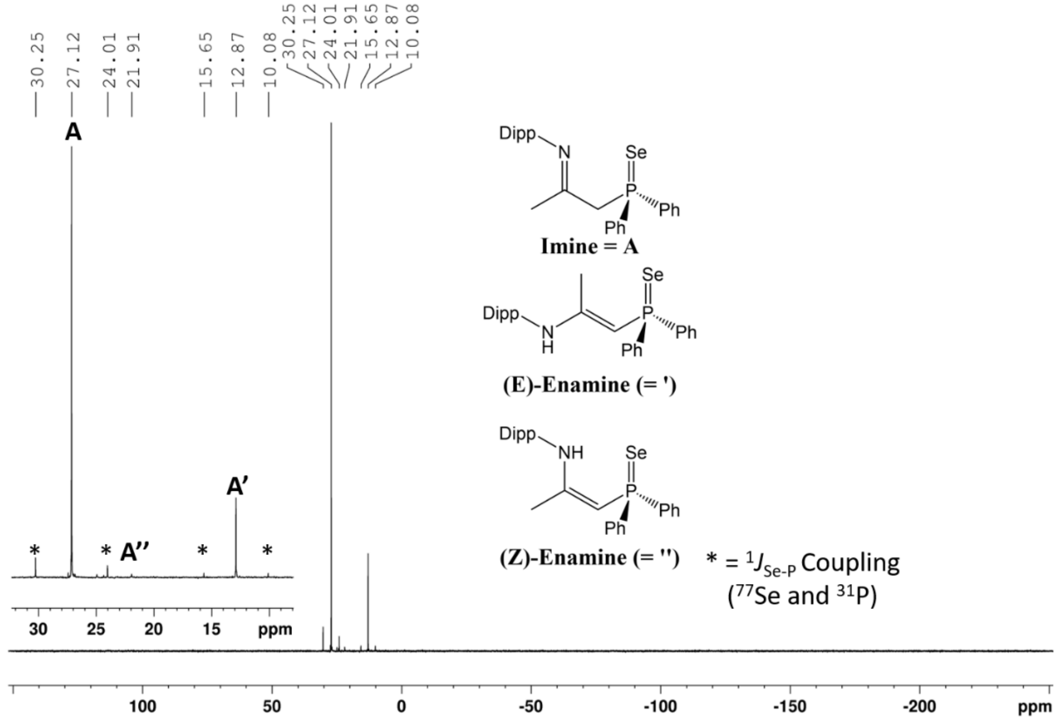


Figure S25 $^{31}P\{^{1}H\}$ NMR spectrum (121 MHz) of compound **4** recorded at 298K in C_6D_6.

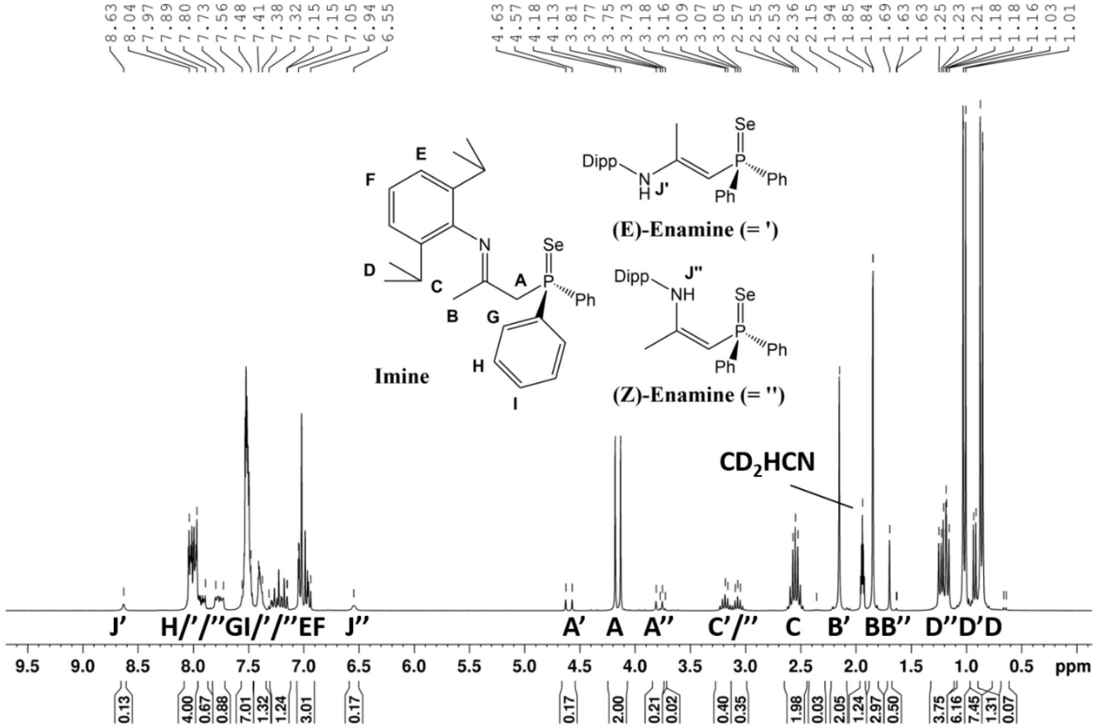


Figure S26 ^{1}H NMR spectrum of compound **4** recorded at 298K in CD_3CN.

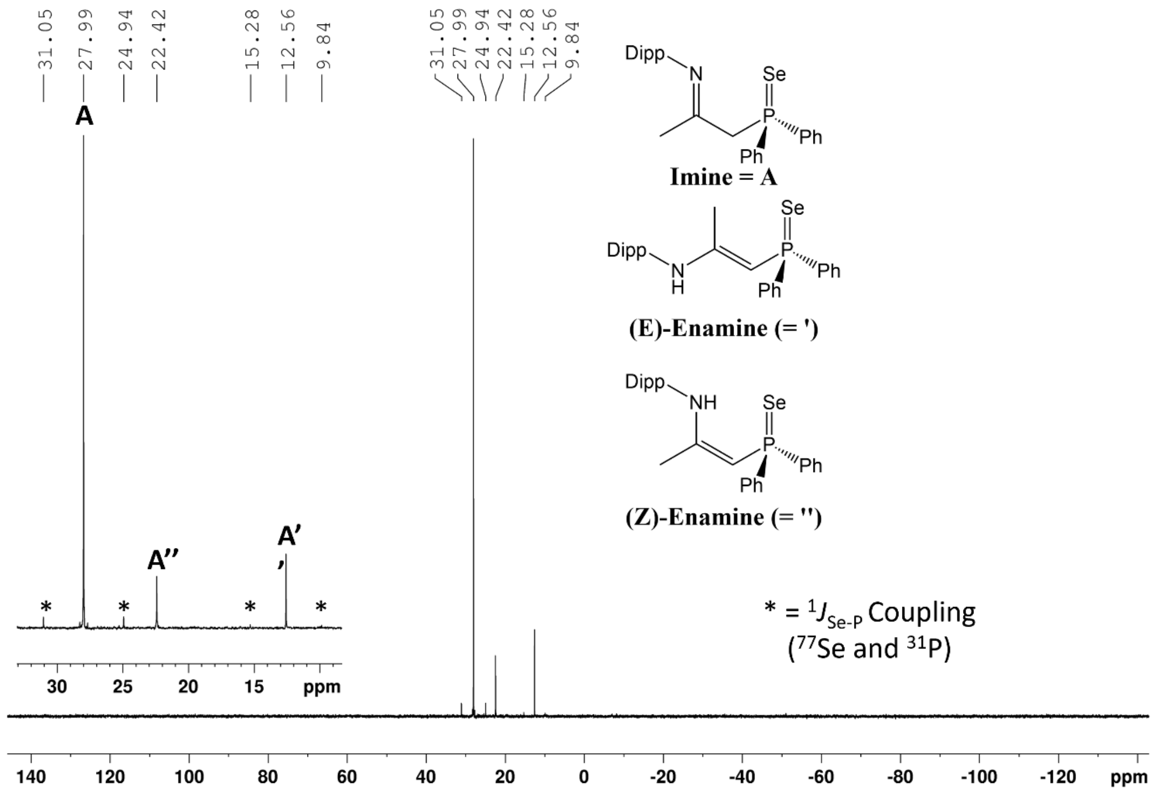


Figure S27 $^{31}P\{^1H\}$ NMR spectrum (121 MHz) of compound **4** recorded at 298K in CD_3CN.

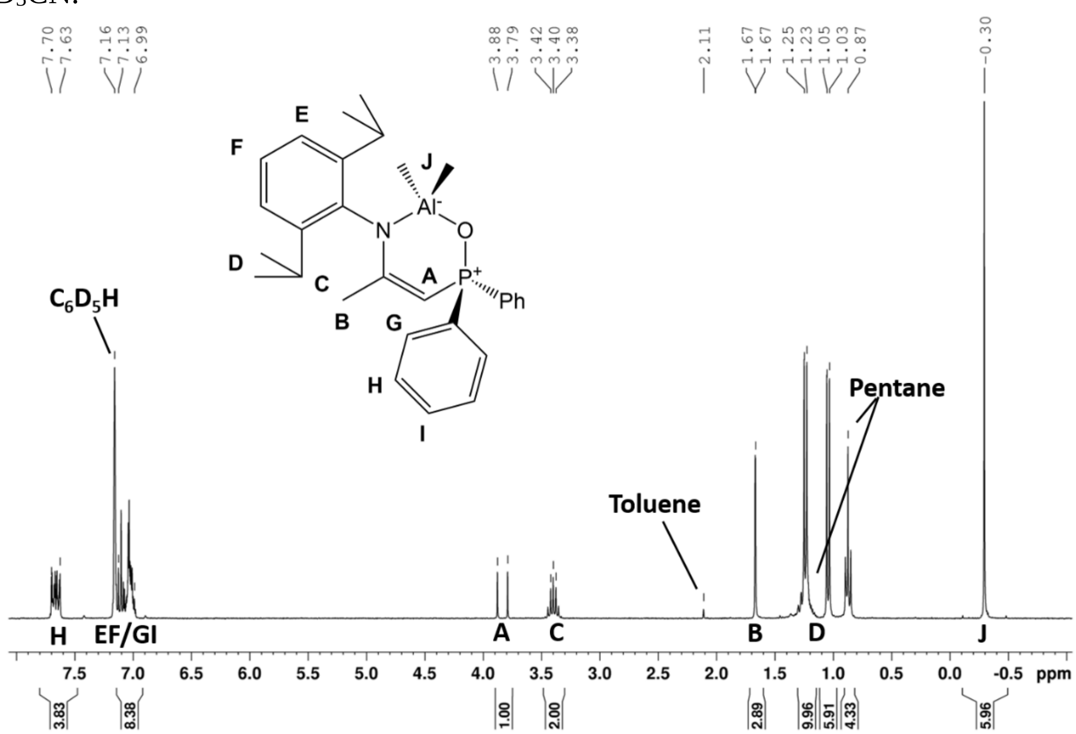


Figure S28 ^1H NMR spectrum of compound **5** recorded at 298K in C_6D_6.

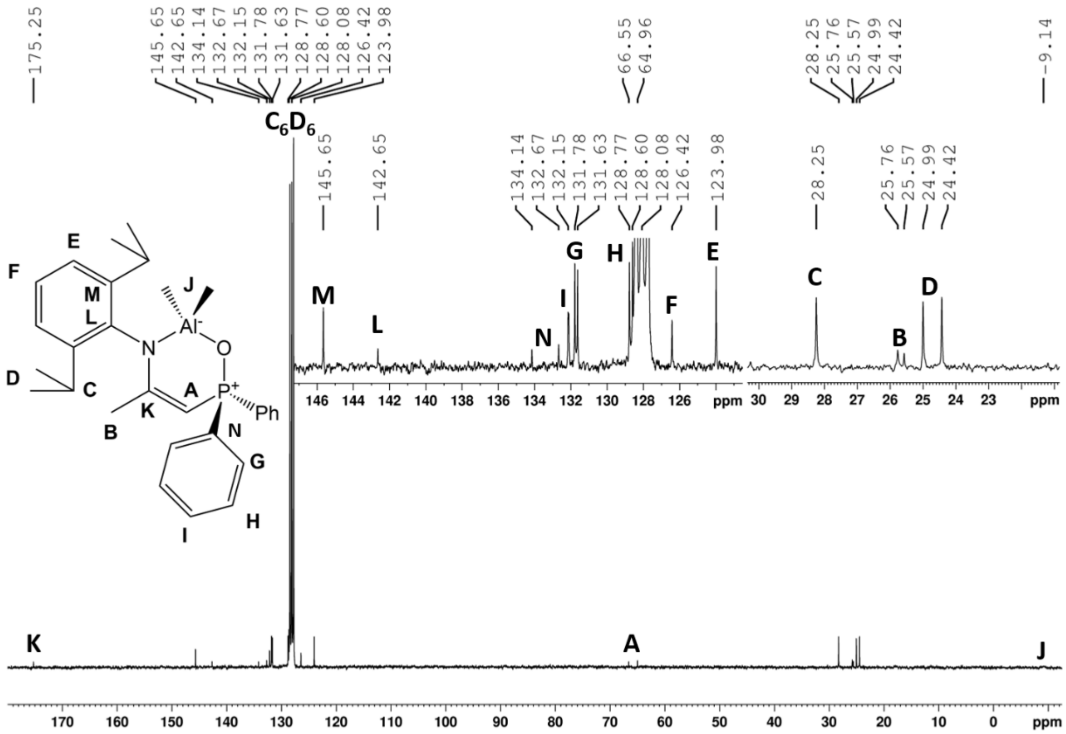


Figure S29 ^{13}C NMR spectrum of compound **5** recorded at 298K in C_6D_6.

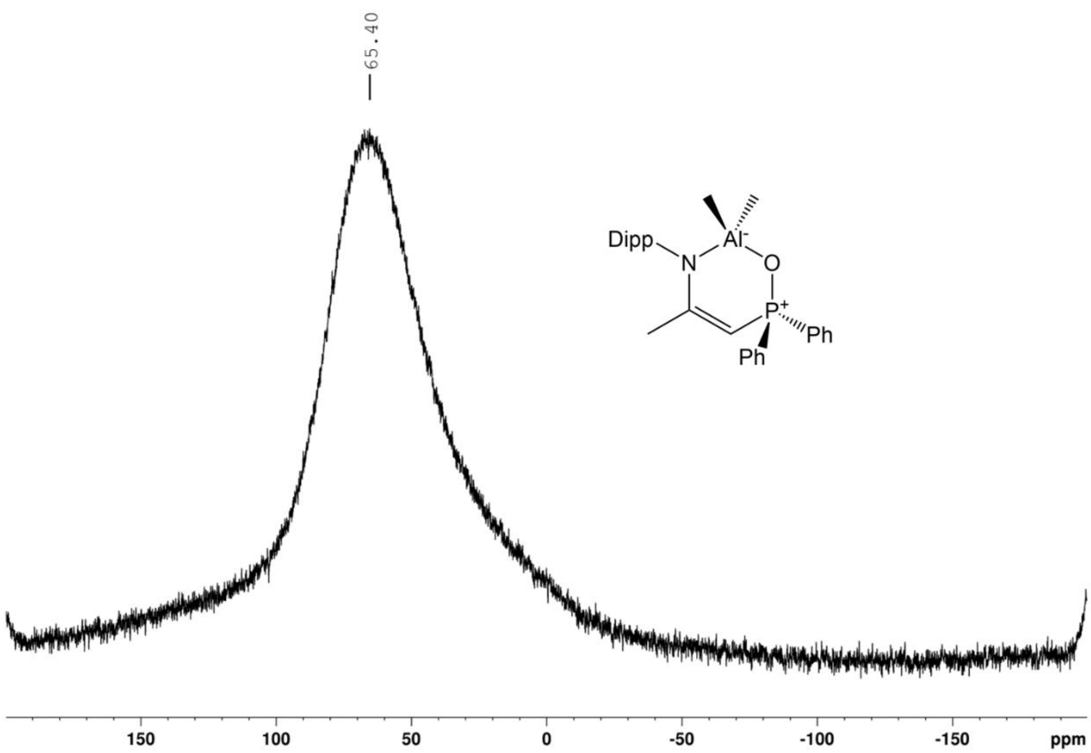


Figure S30 ^{27}Al NMR spectrum of compound **5** recorded at 298K in C_6D_6.

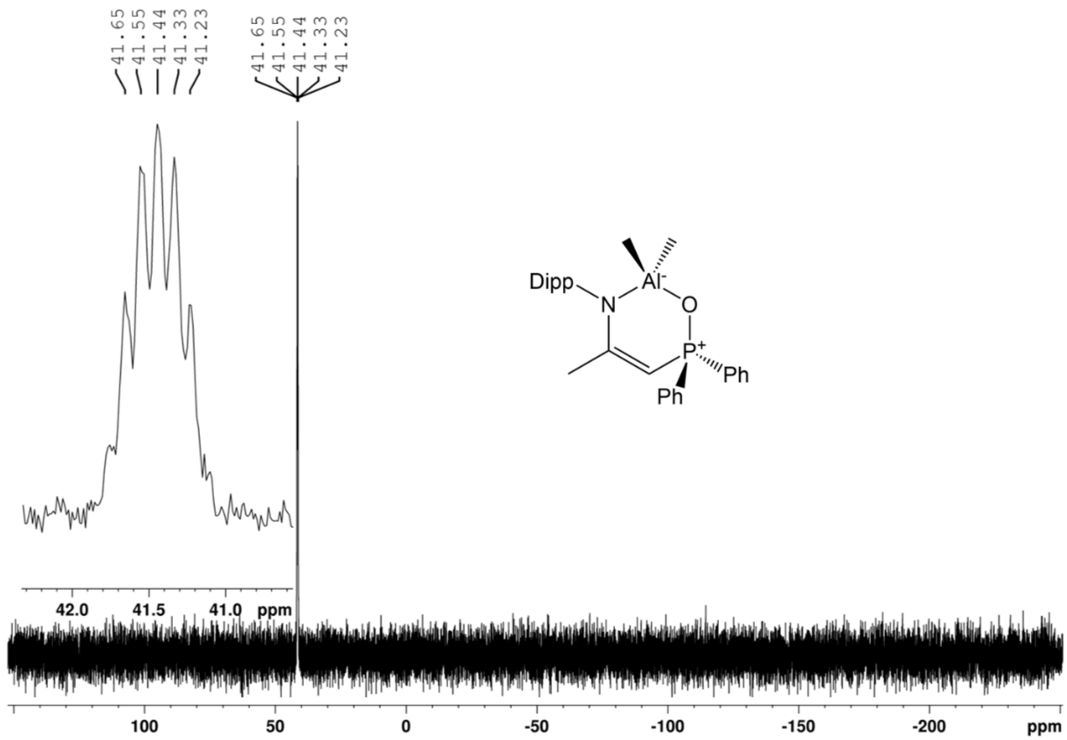


Figure S31 ^{31}P NMR spectrum of compound **5** recorded at 298K in C_6D_6.

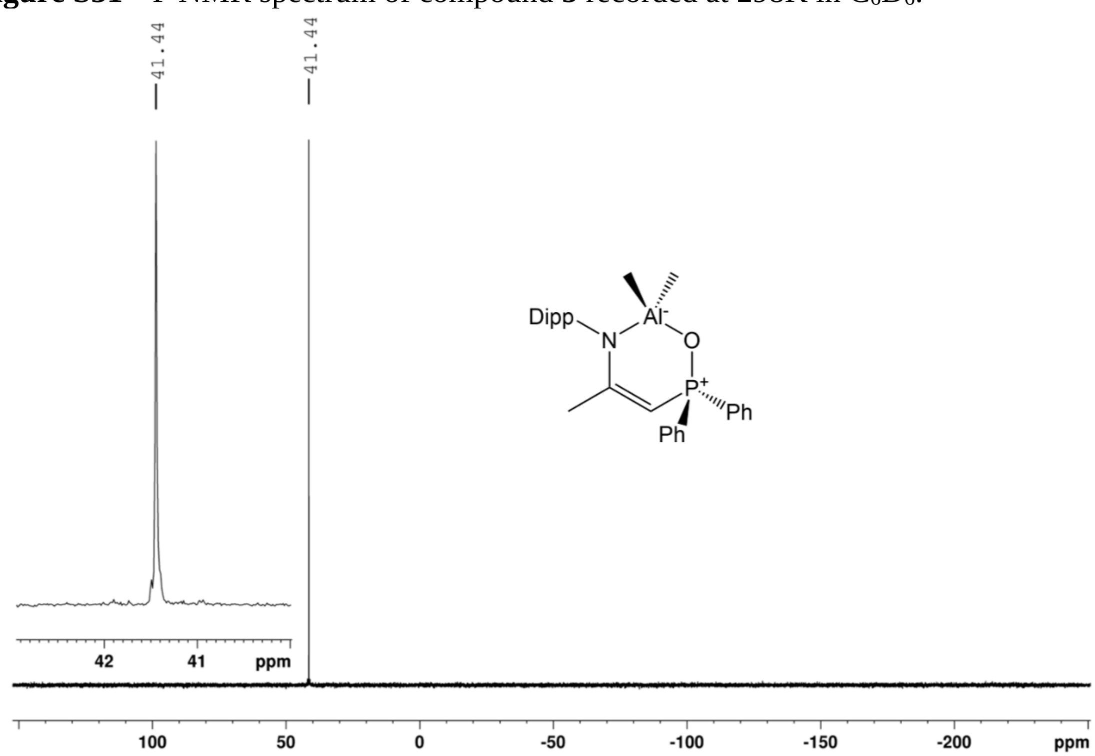


Figure S32 $^{31}P\{^1H\}$ NMR spectrum of compound **5** recorded at 298K in C_6D_6.

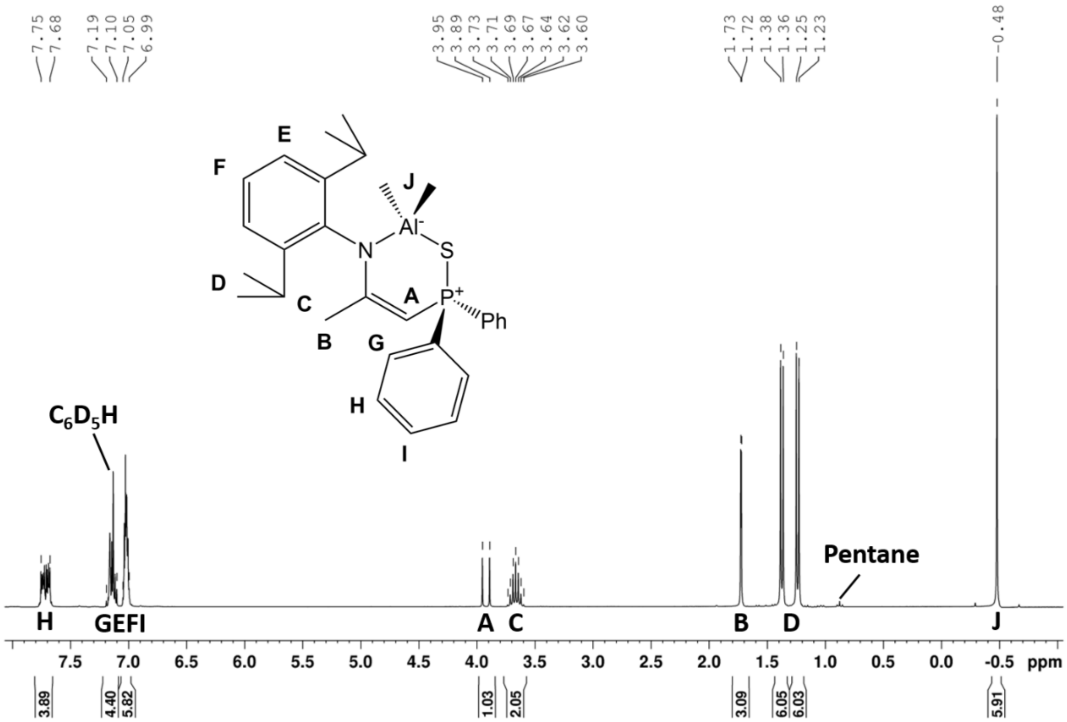


Figure S33 ^{1}H NMR spectrum of compound **6** recorded at 298K in C_6D_6.

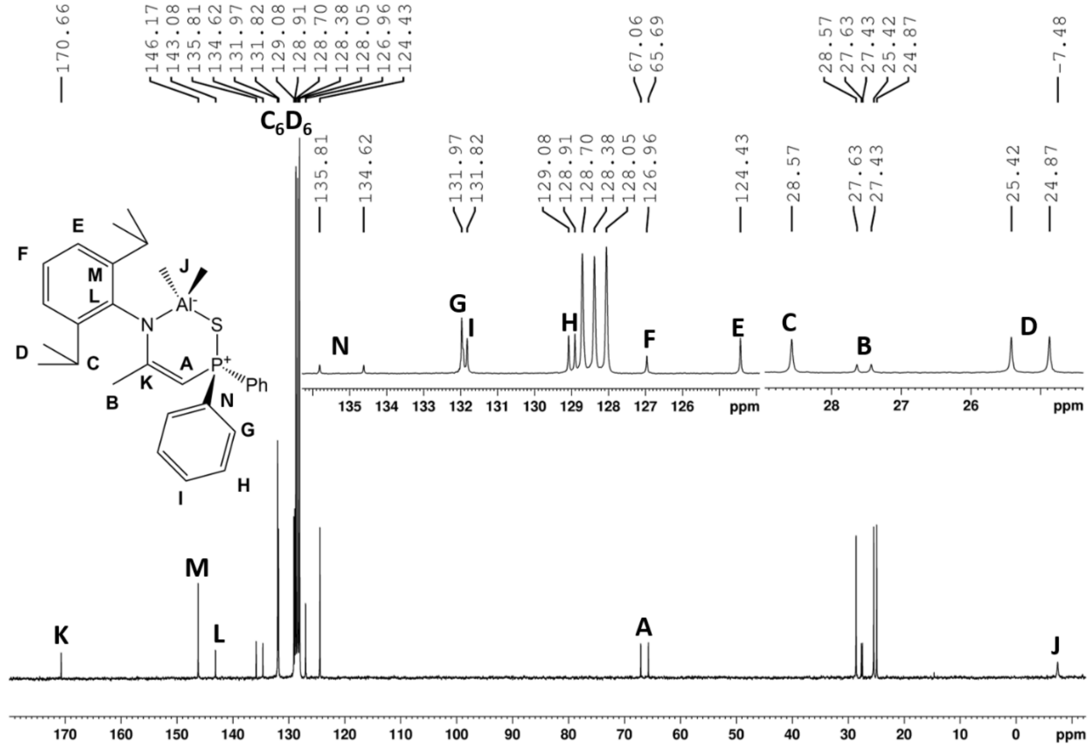


Figure S34 ^{13}C NMR spectrum of compound **6** recorded at 298K in C_6D_6.

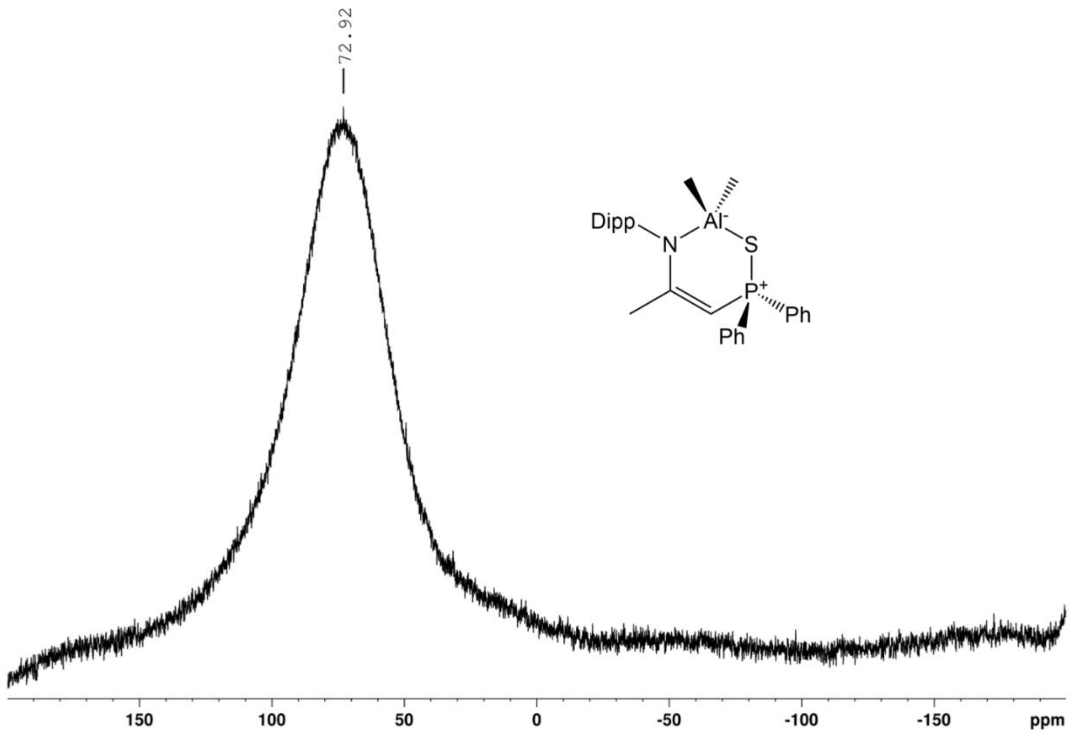


Figure S35 ^{27}Al NMR spectrum of compound **6** recorded at 298K C_6D_6.

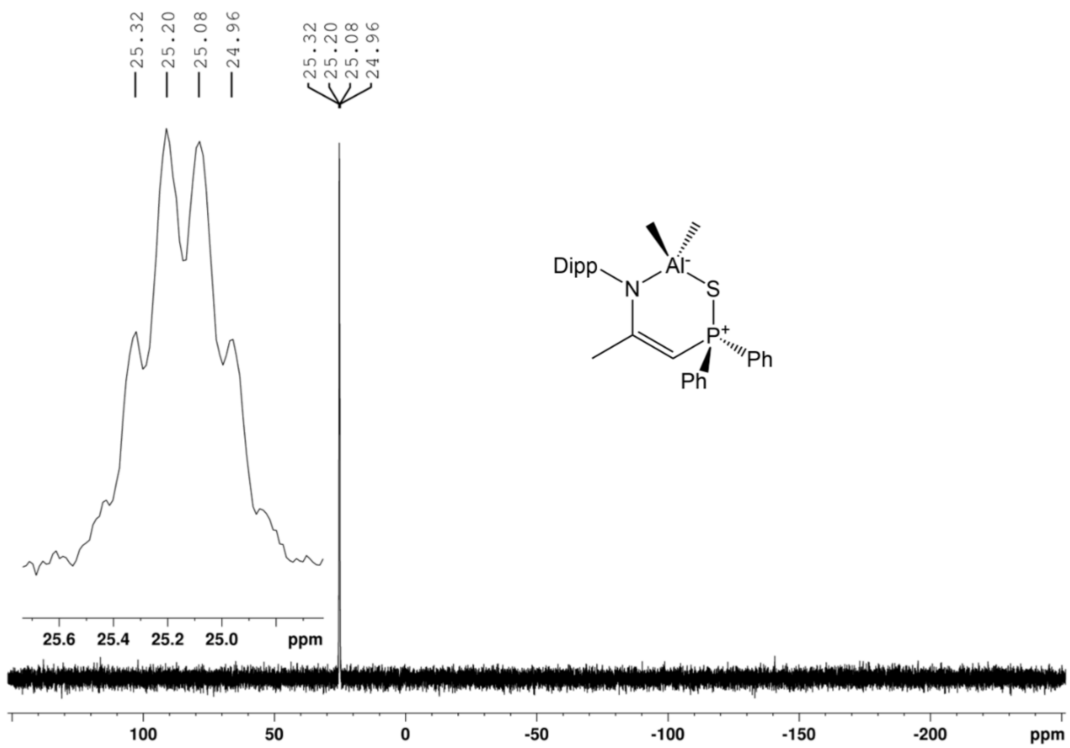


Figure S36 ^{31}P NMR spectrum of compound **6** recorded at 298K in C_6D_6.

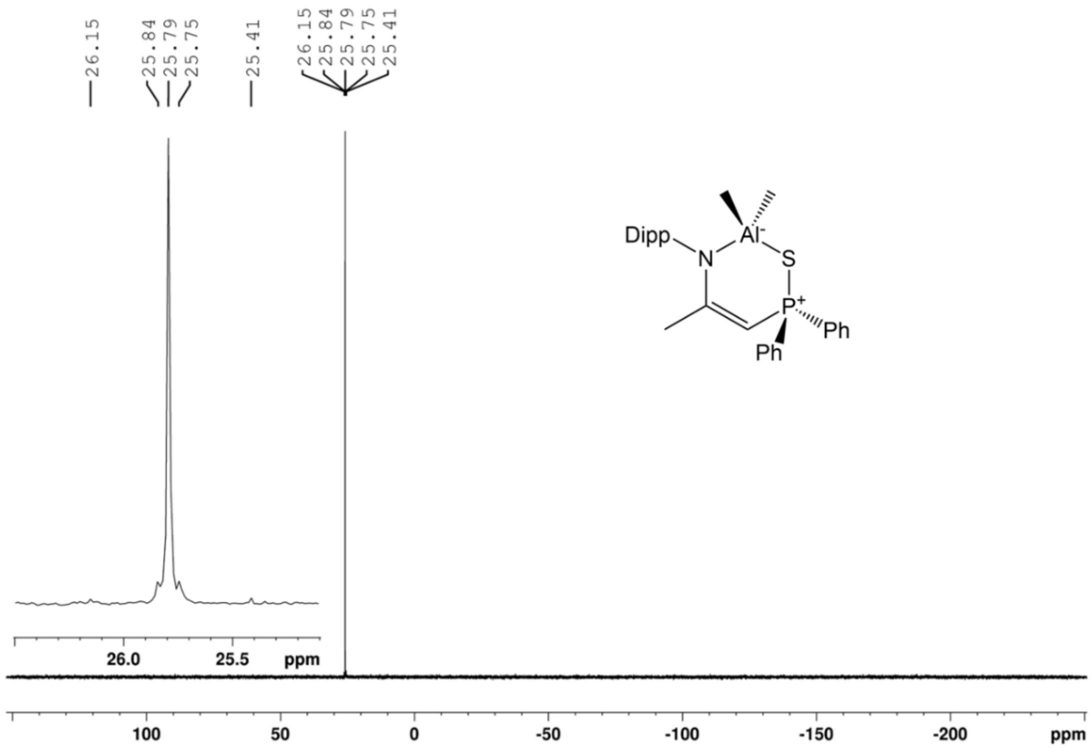


Figure S37 $^{31}P\{^{1}H\}$ NMR spectrum of compound **6** recorded at 298K in C_6D_6.

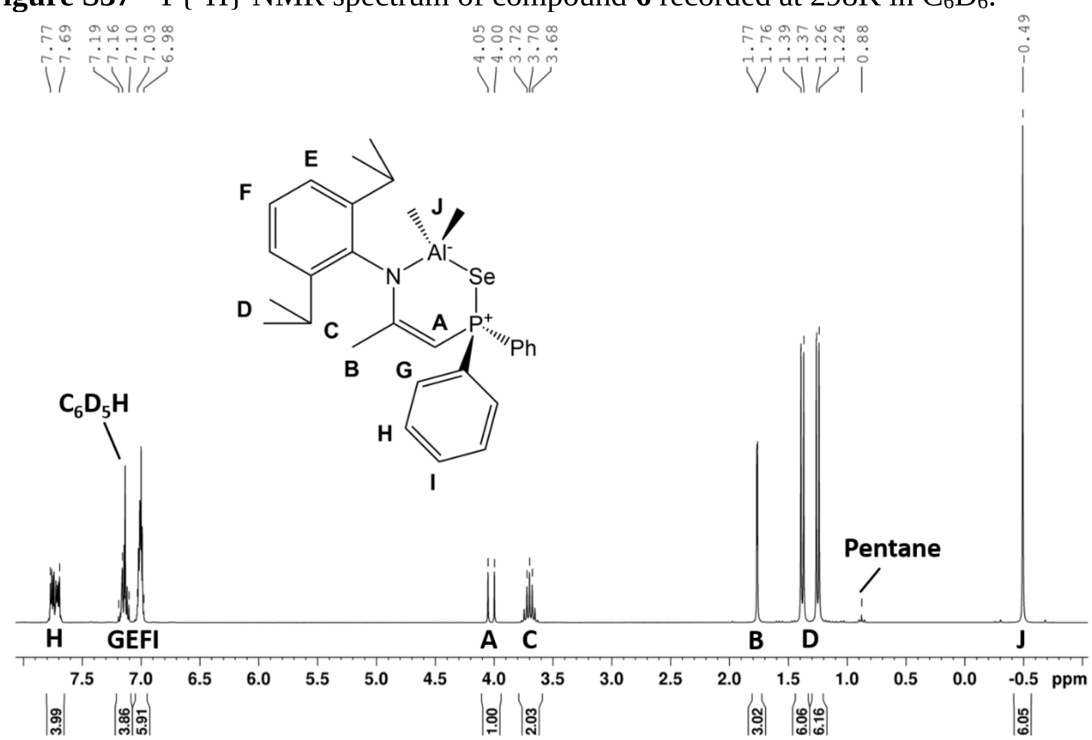


Figure S38 ^{1}H NMR spectrum of compound **7** recorded at 298K in C_6D_6.

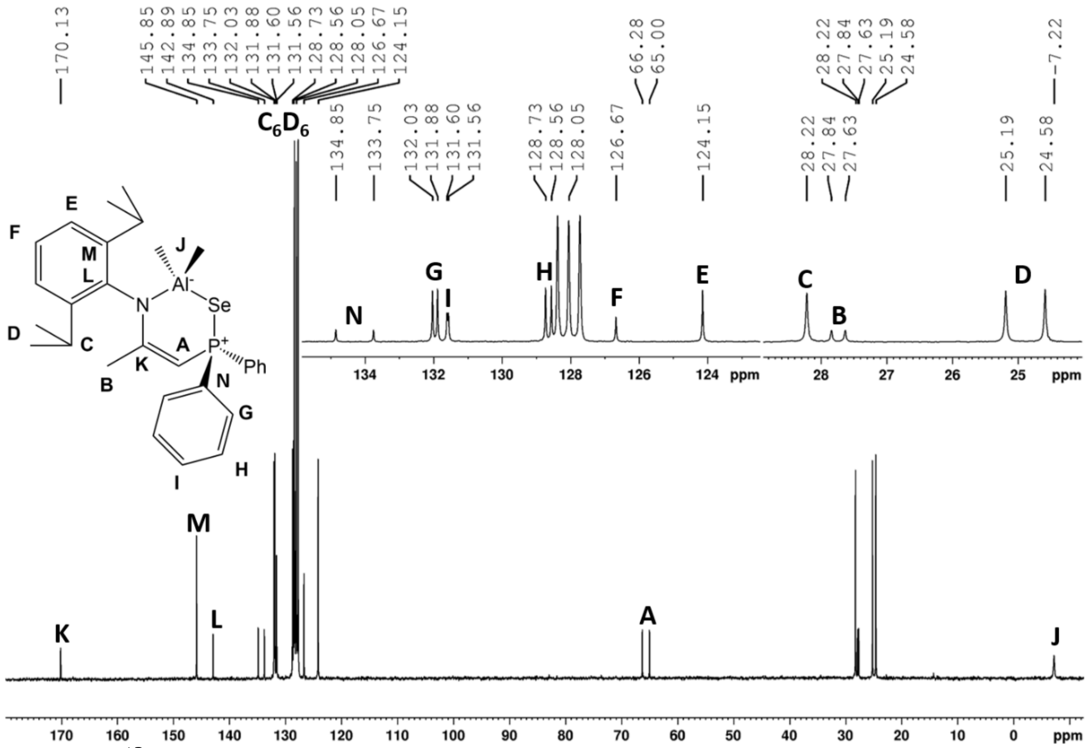


Figure S39 ^{13}C NMR spectrum of compound **7** recorded at 298K in C_6D_6.

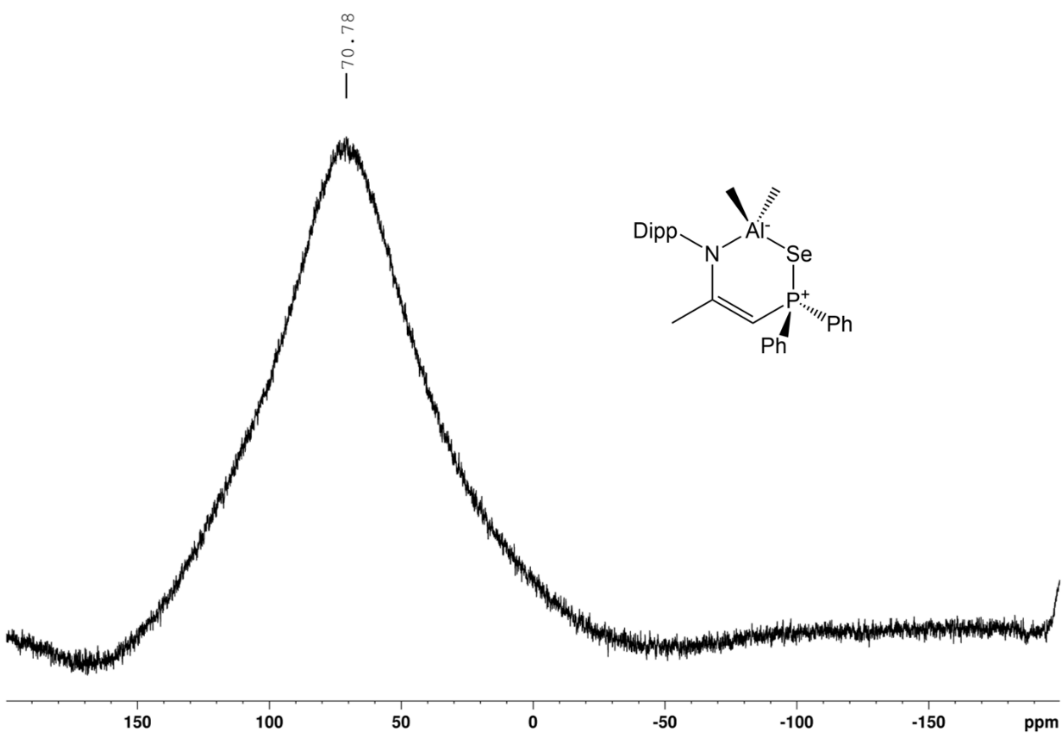


Figure S40 ^{27}Al NMR spectrum of compound **7** recorded at 298K in C_6D_6.

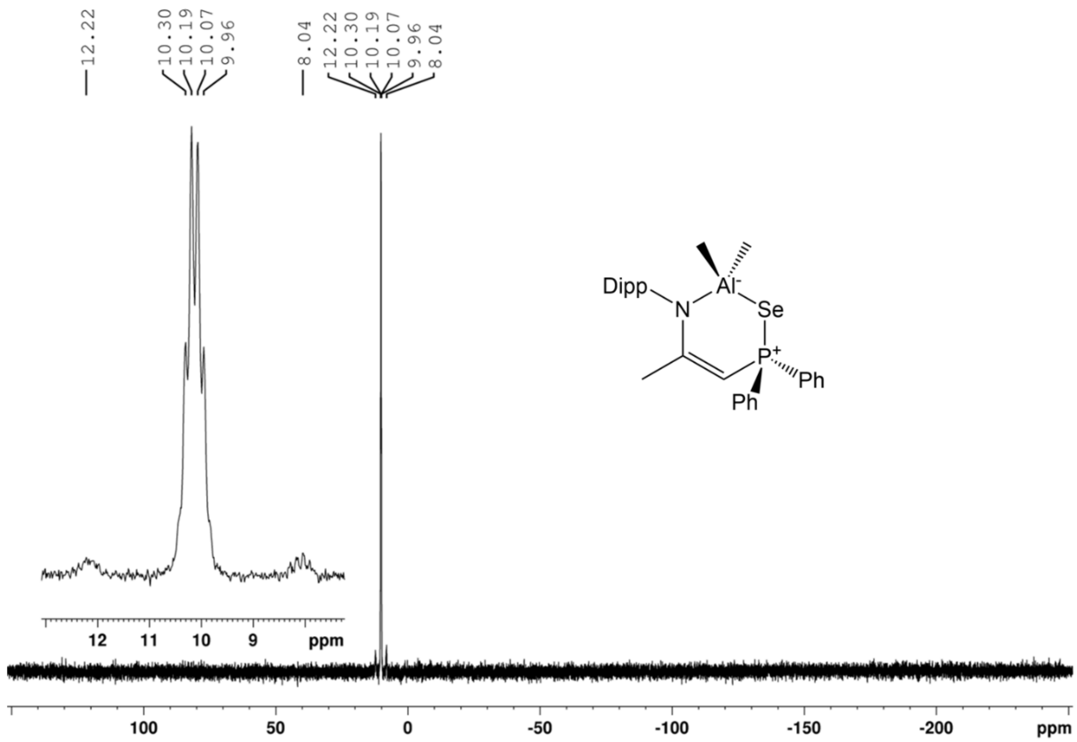


Figure S41 ^{31}P NMR spectrum of compound **7** recorded at 298K in C_6D_6.

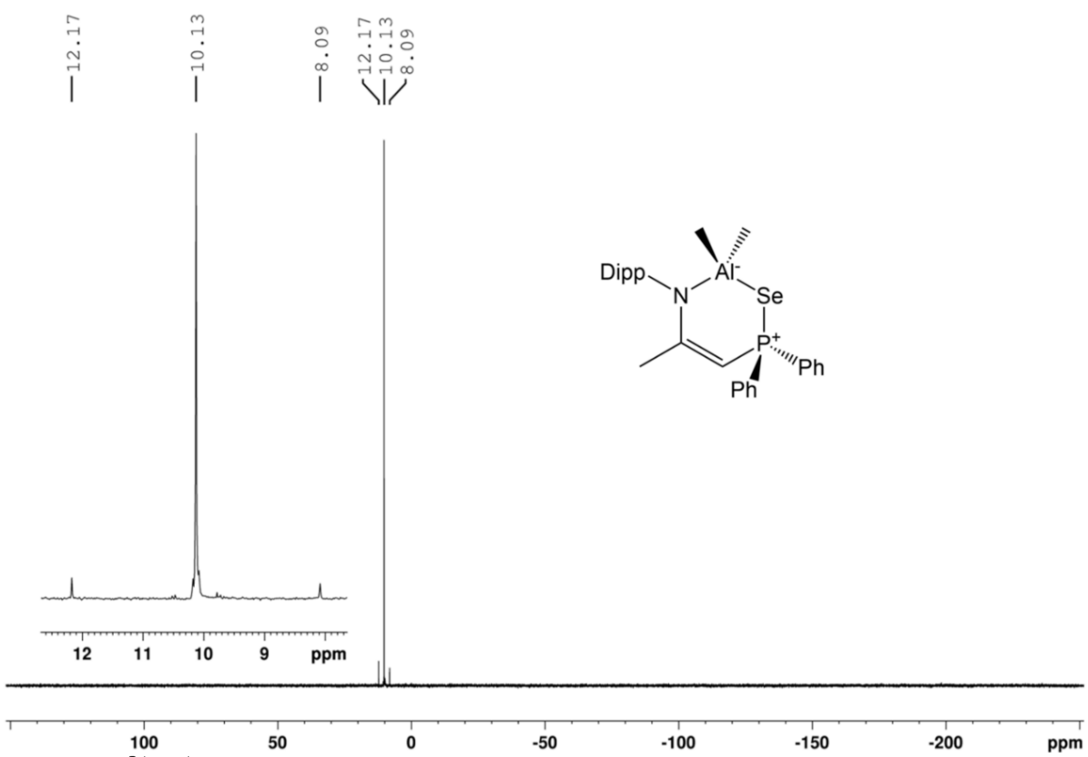


Figure S42 $^{31}P\{^{1}H\}$ NMR spectrum of compound **7** recorded at in C_6D_6.

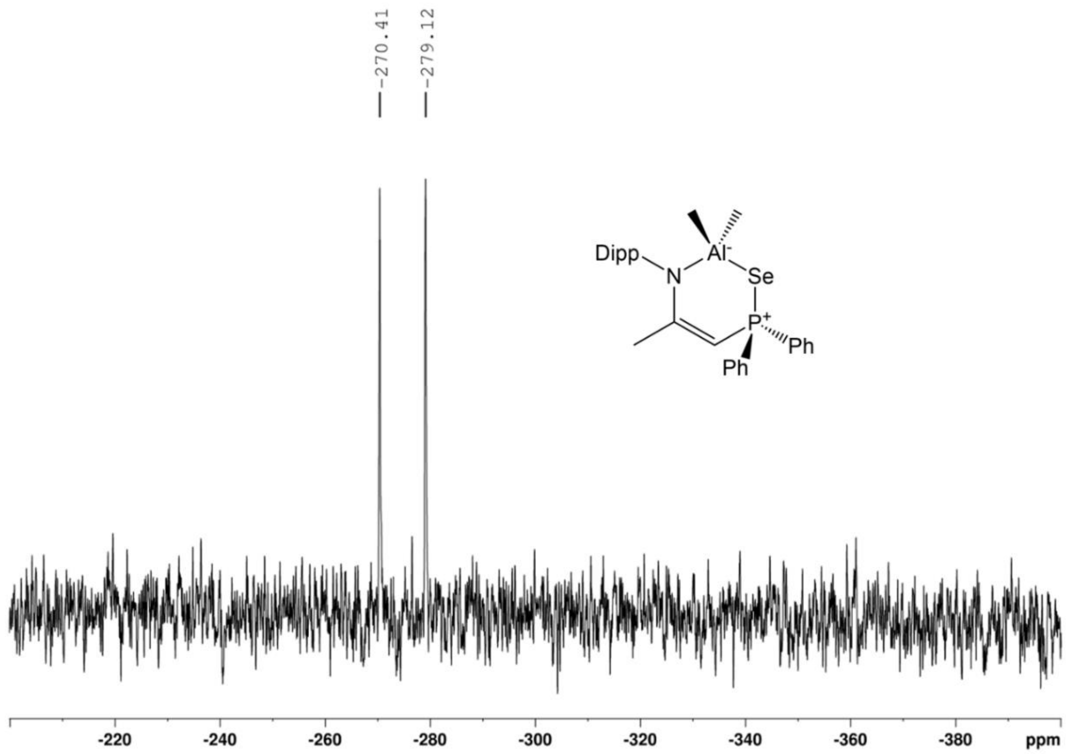


Figure S43 ^{77}Se NMR spectrum of compound **7** recorded at 298K in C_6D_6.

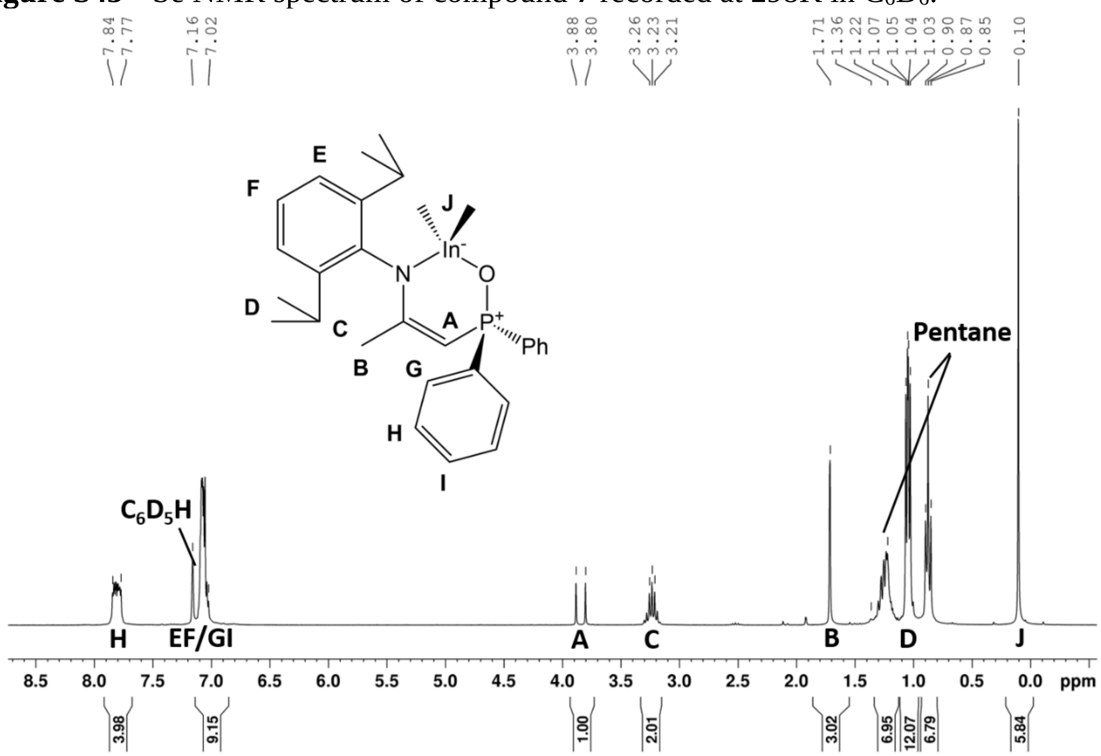


Figure S44 ^{1}H NMR spectrum of compound **8** recorded at 298K in C_6D_6.

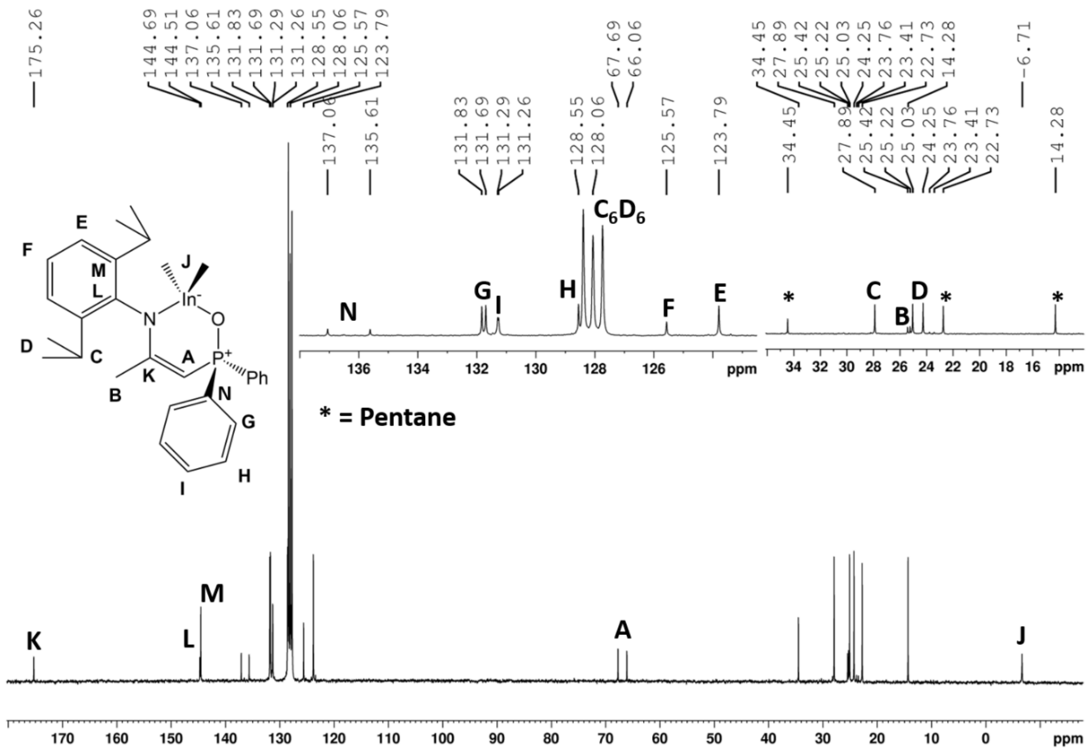


Figure S45 ^{13}C NMR spectrum of compound **8** recorded at 298K in C_6D_6.

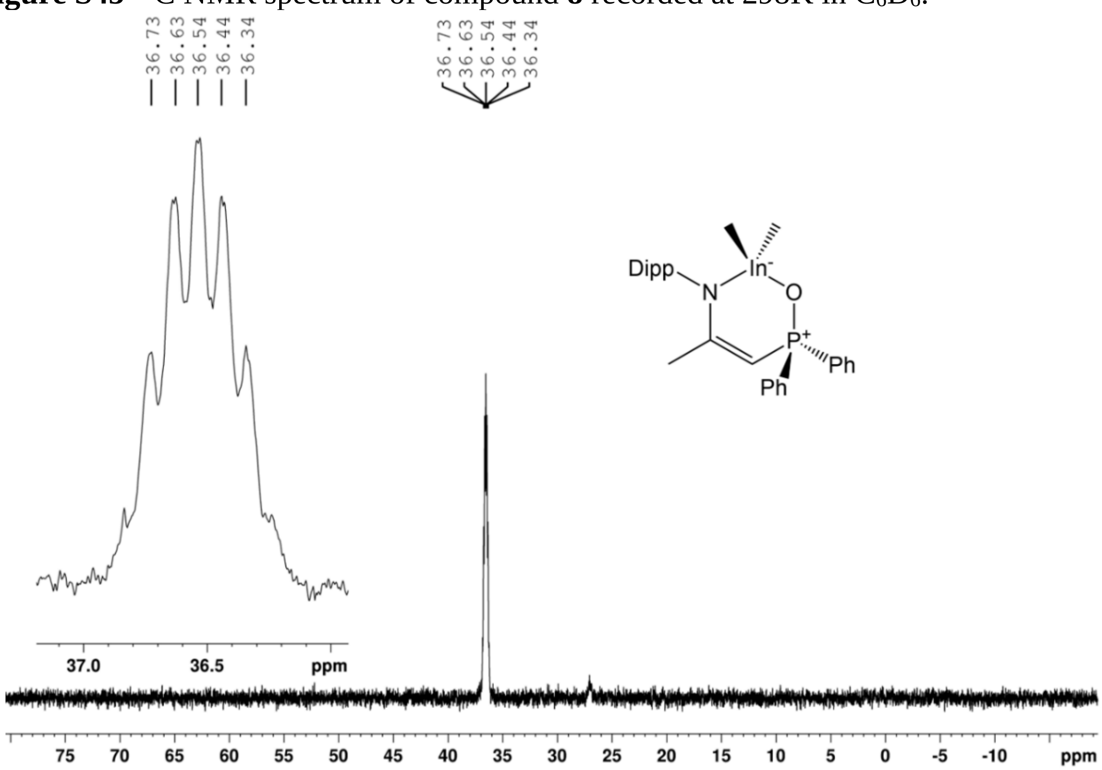


Figure S46 ^{31}P NMR spectrum of compound **8** recorded at 298K in C_6D_6.

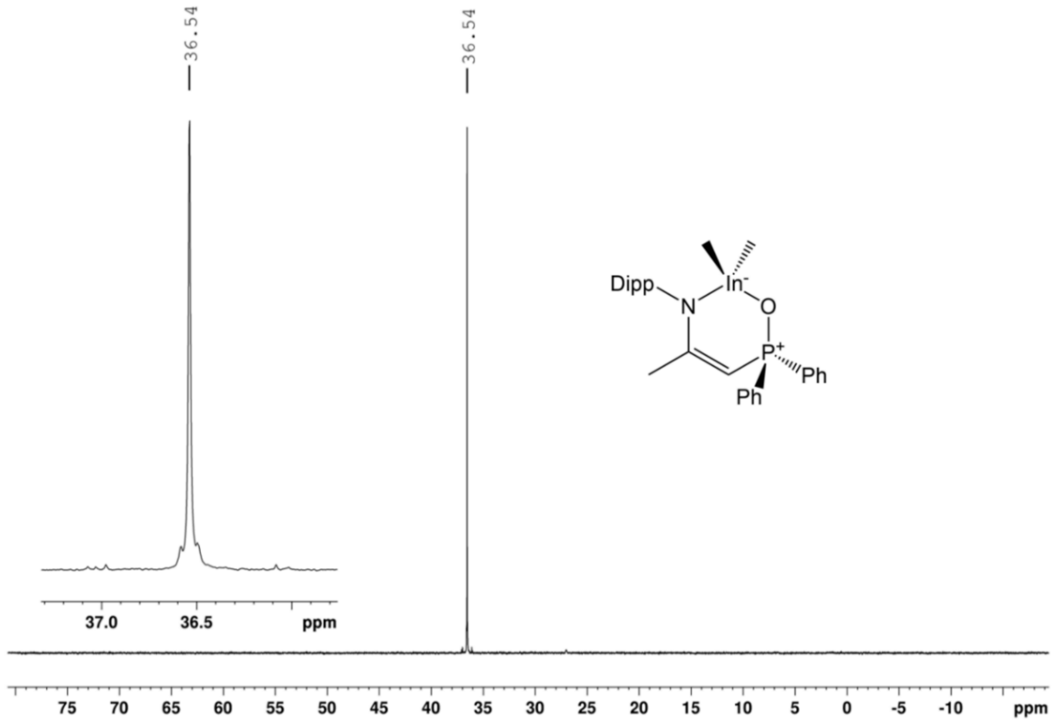


Figure S47 $^{31}P\{^1H\}$ NMR spectrum of compound **8** recorded at 298K in C_6D_6.

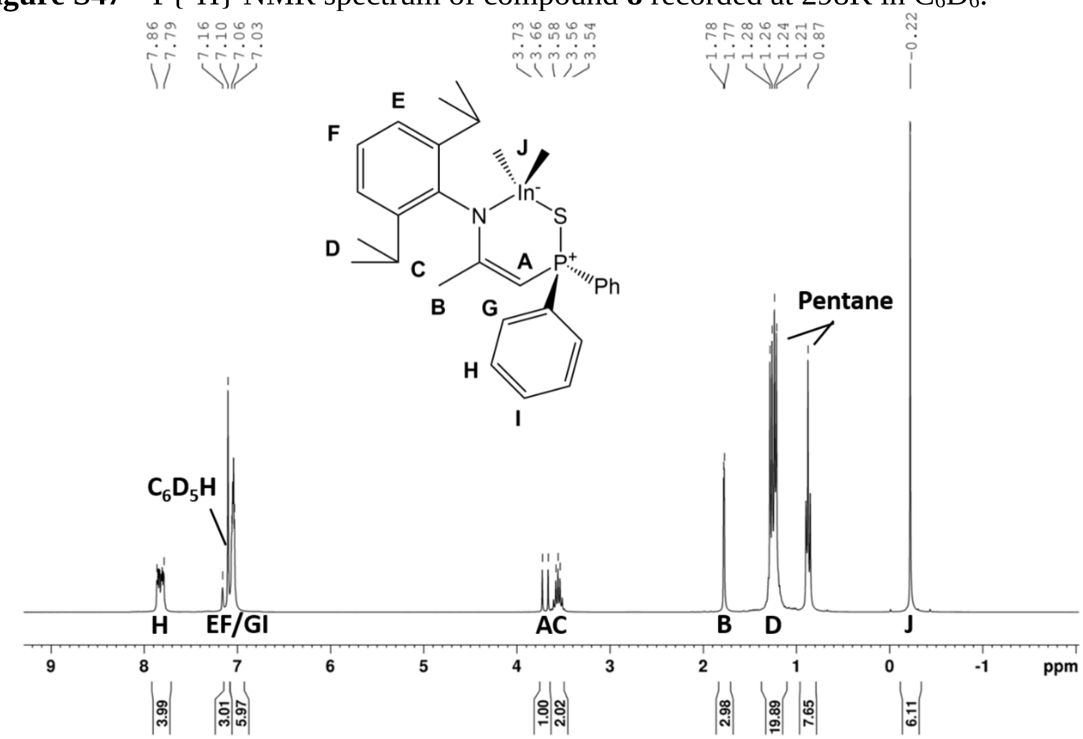


Figure S48 ^1H NMR spectrum of compound **9** recorded at 298K in C_6D_6.

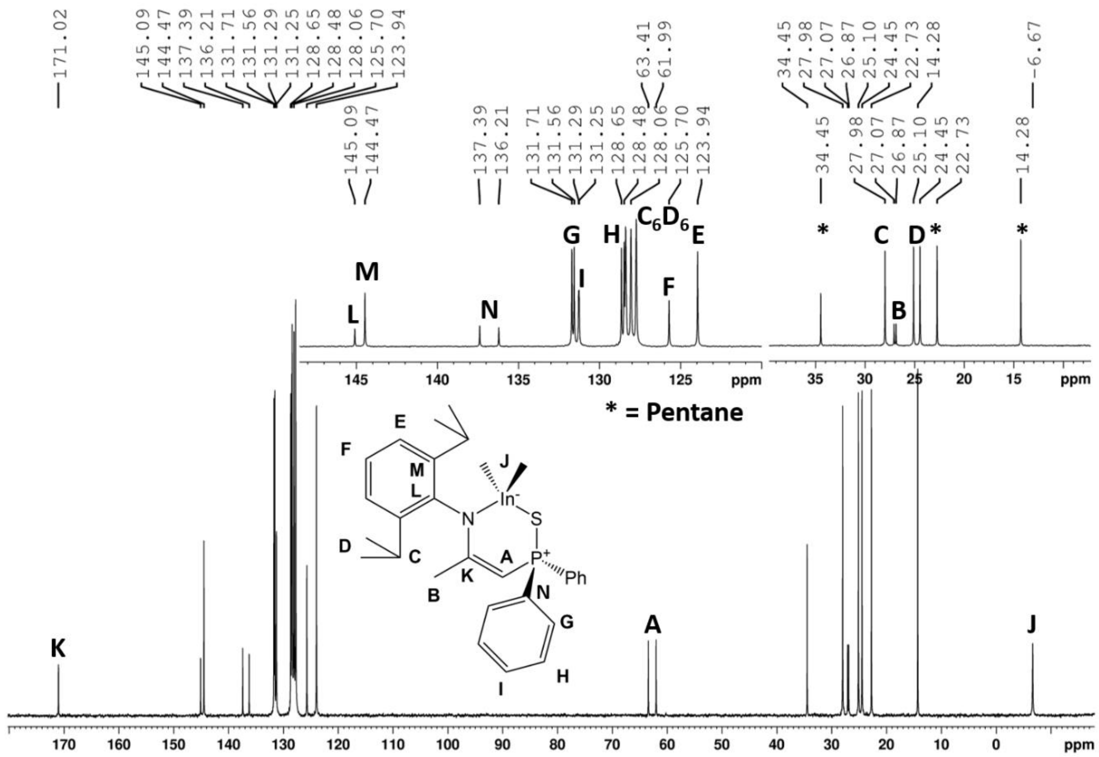


Figure S49 ^{13}C NMR spectrum of compound **9** recorded at 298K in C_6D_6.

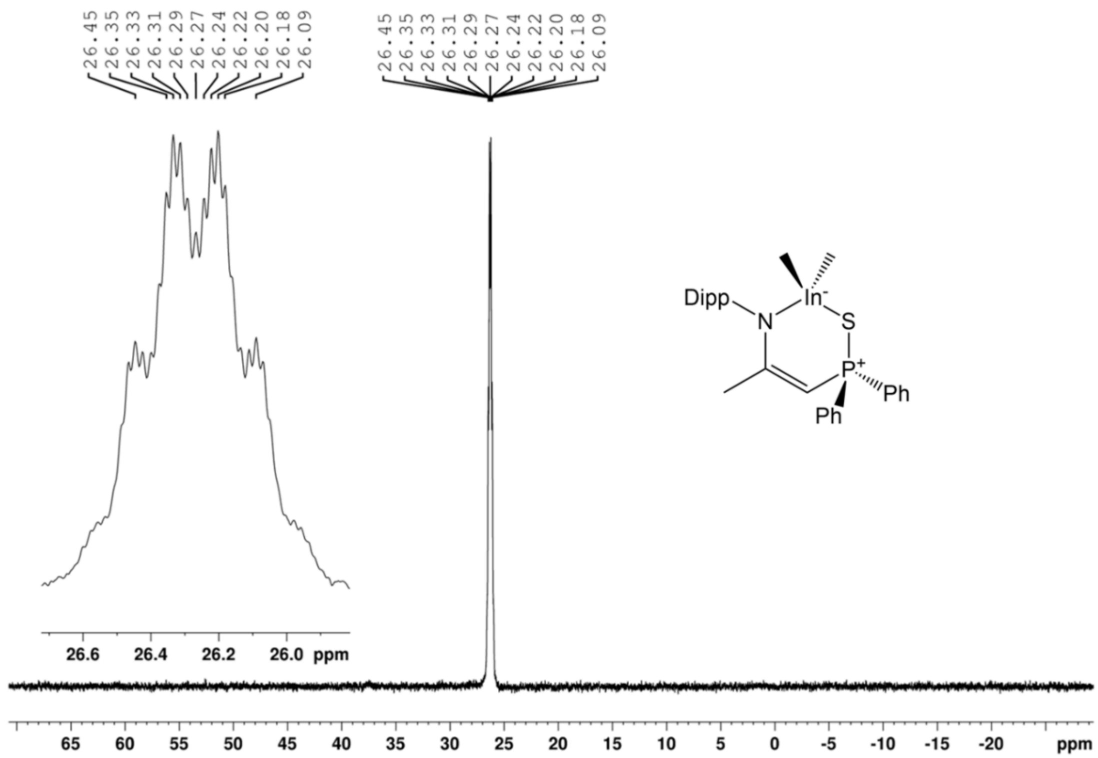


Figure S50 ^{31}P NMR spectrum of compound **9** recorded at 298K in C_6D_6.

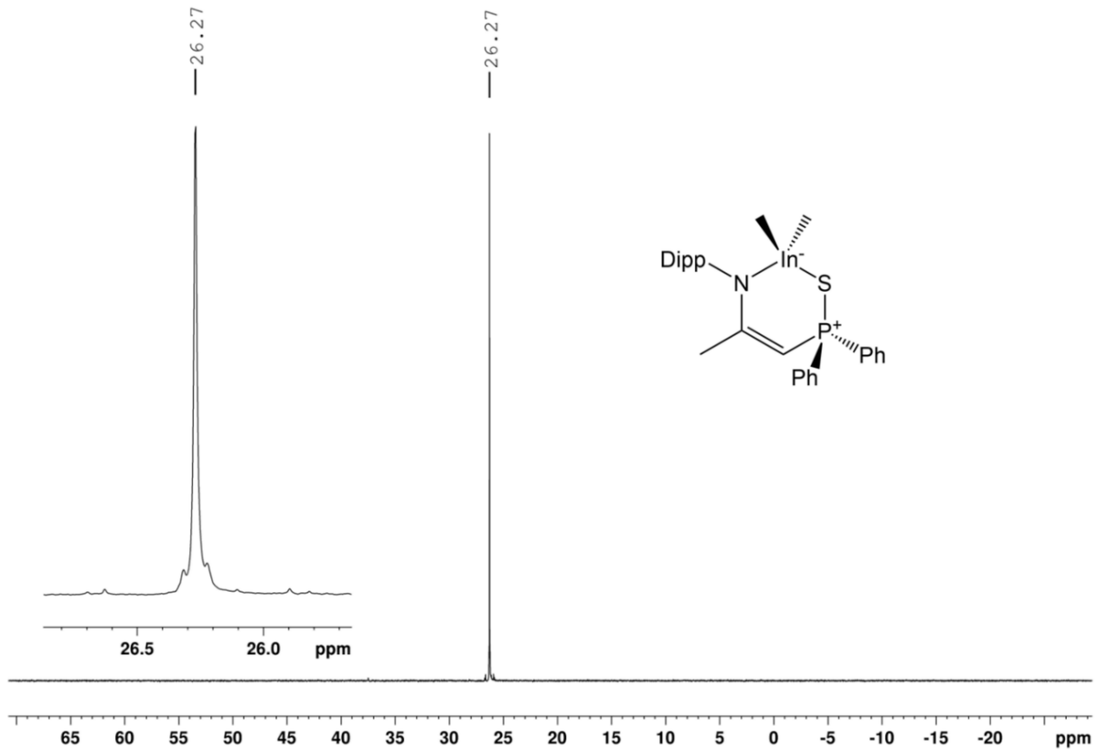


Figure S51 $^{31}P\{^{1}H\}$ NMR spectrum of compound **9** recorded at 298K in C_6D_6.

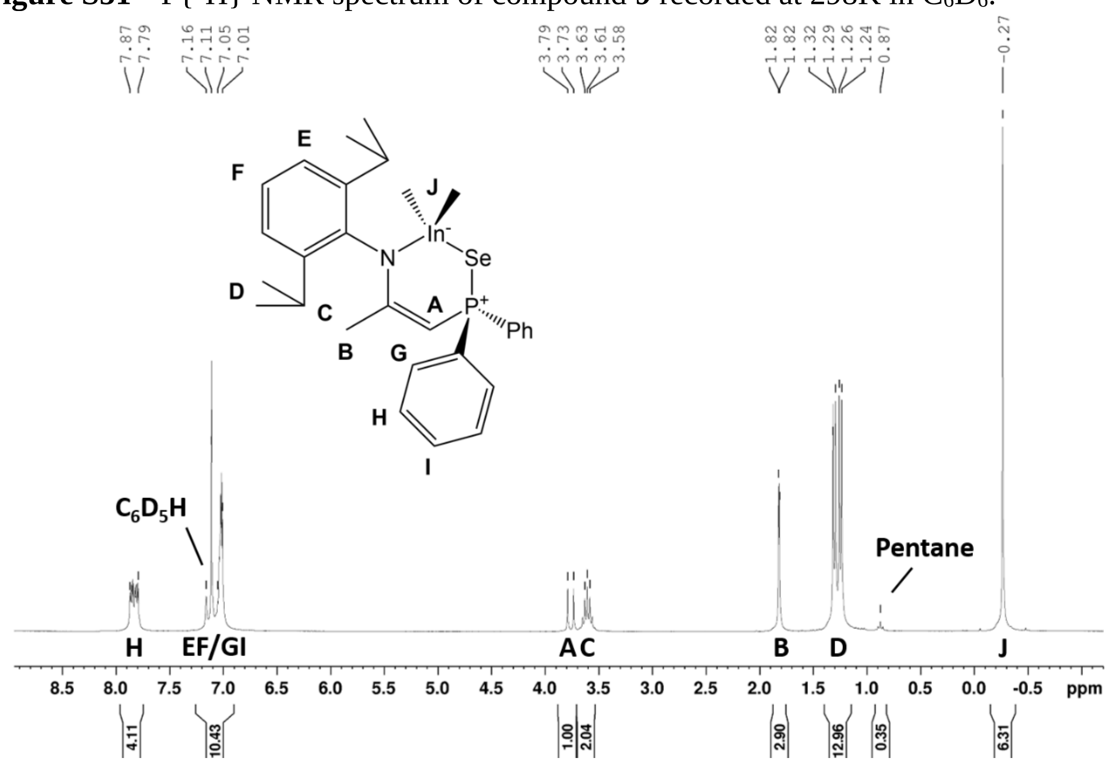


Figure S52 ^{1}H NMR spectrum of compound **10** recorded at 298K in C_6D_6.

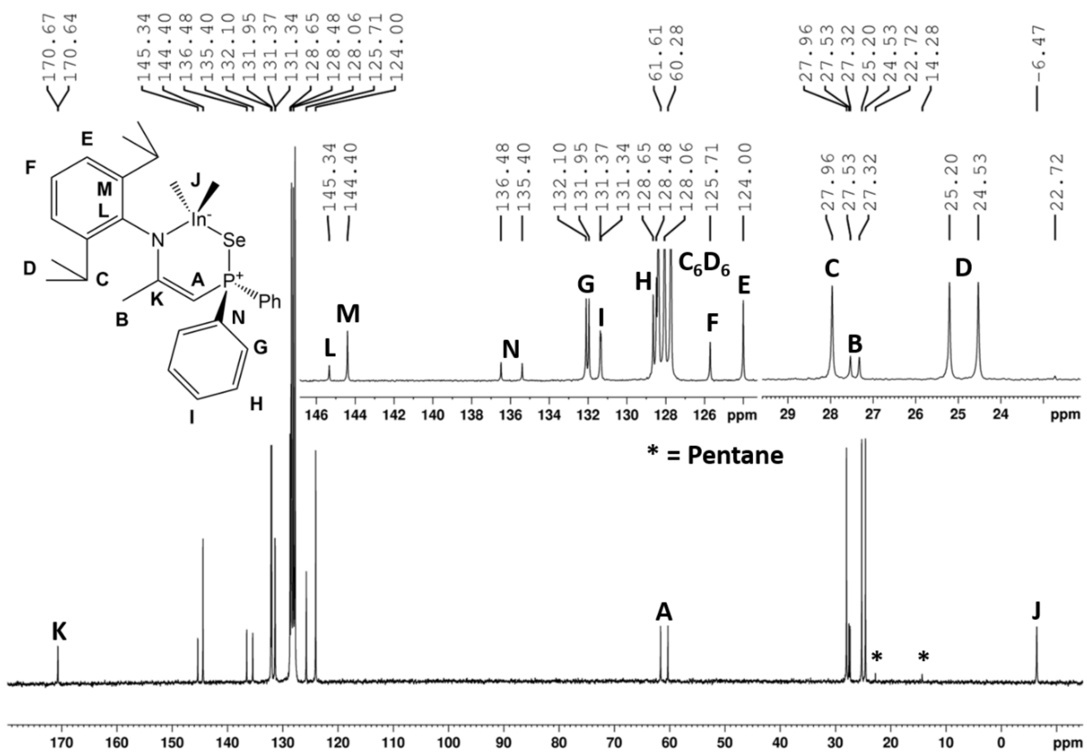


Figure S53 ^{13}C NMR spectrum of compound **10** recorded at 298K in C_6D_6.

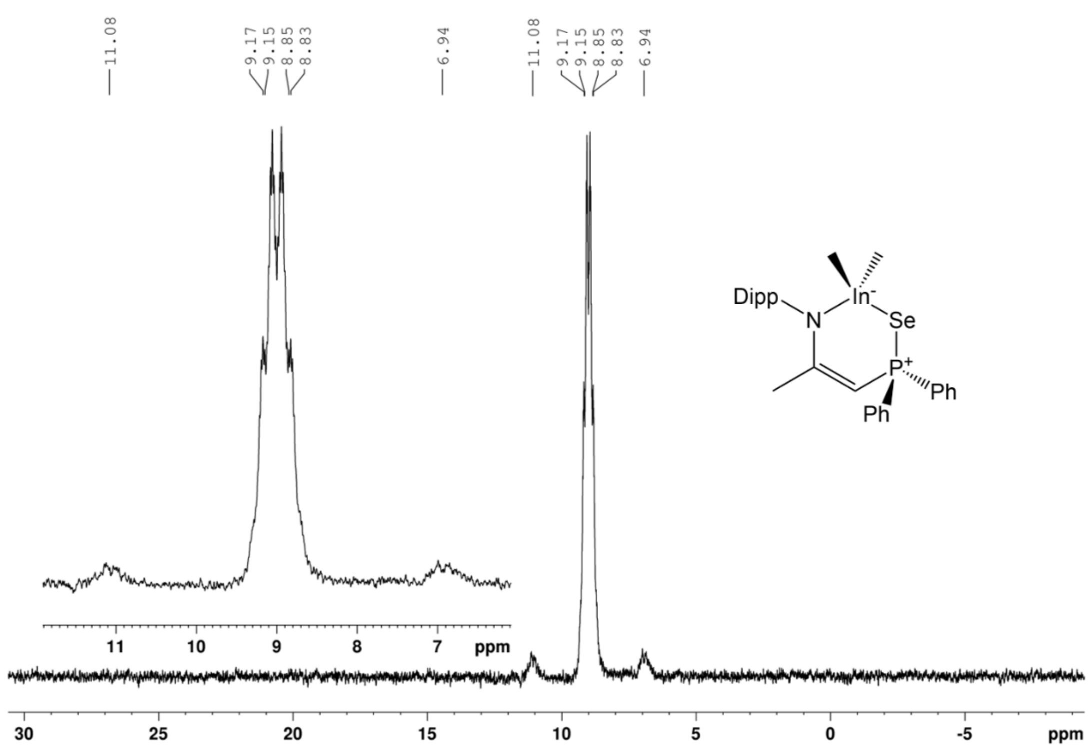


Figure S54 ^{31}P NMR spectrum of compound **10** recorded at 298K in C_6D_6.

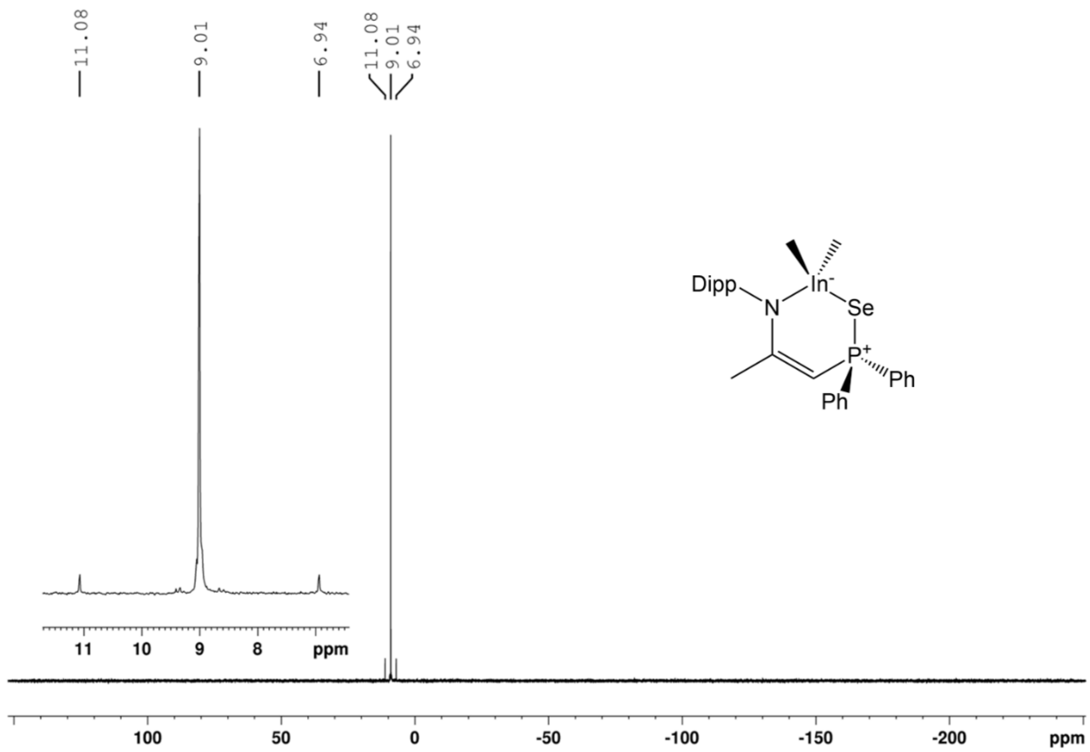


Figure S55 $^{31}P\{^{1}H\}$ NMR spectrum of compound **10** recorded at in C_6D_6.

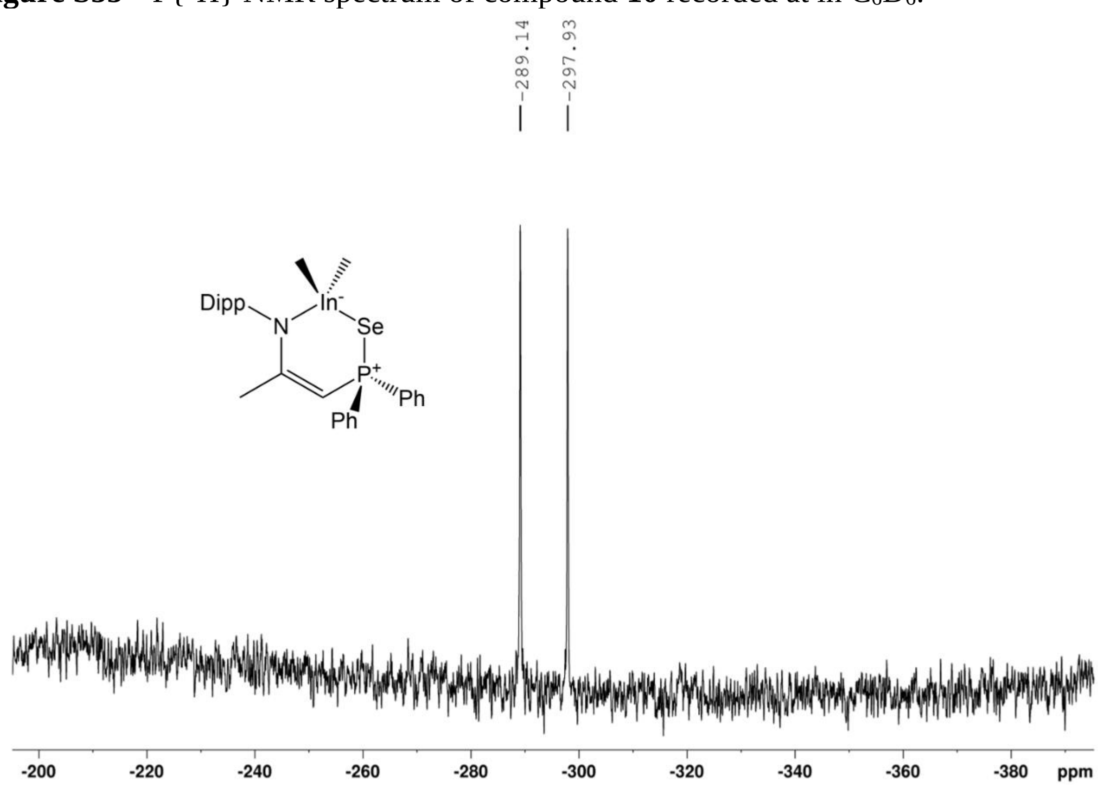


Figure S56 ^{77}Se NMR spectrum of compound **10** recorded at 298K in C_6D_6.

3 - Infrared Spectroscopy

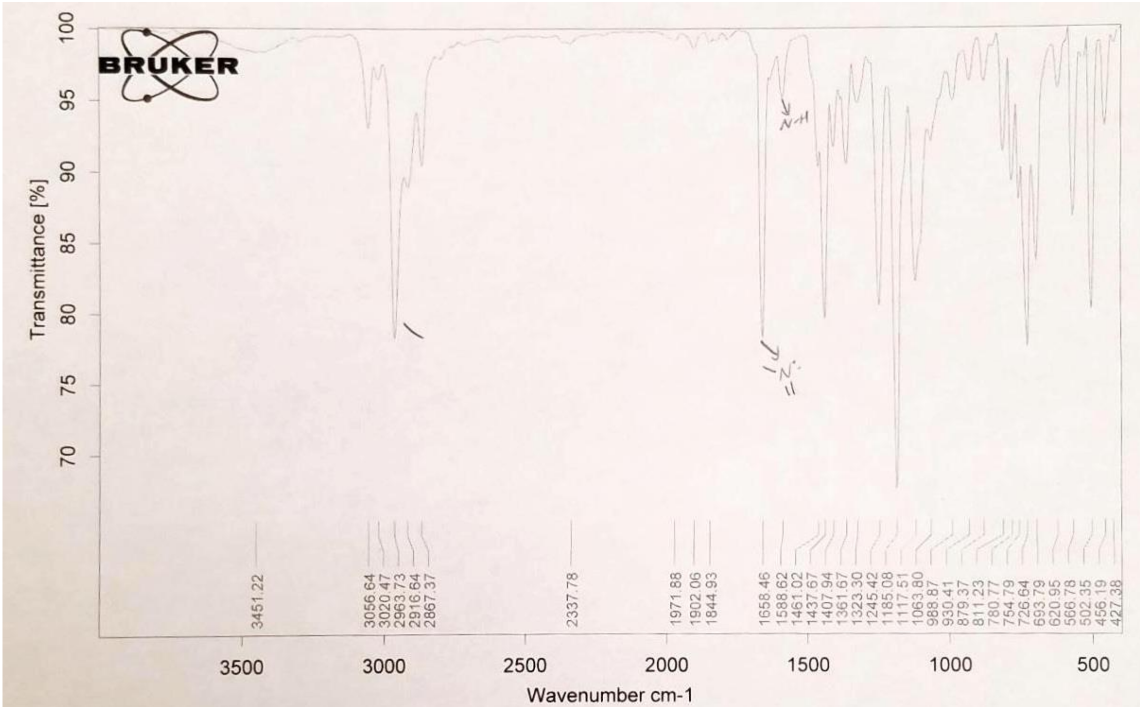


Figure S57 Infrared spectrum of compound **2a** as a pressed KBr pellet.

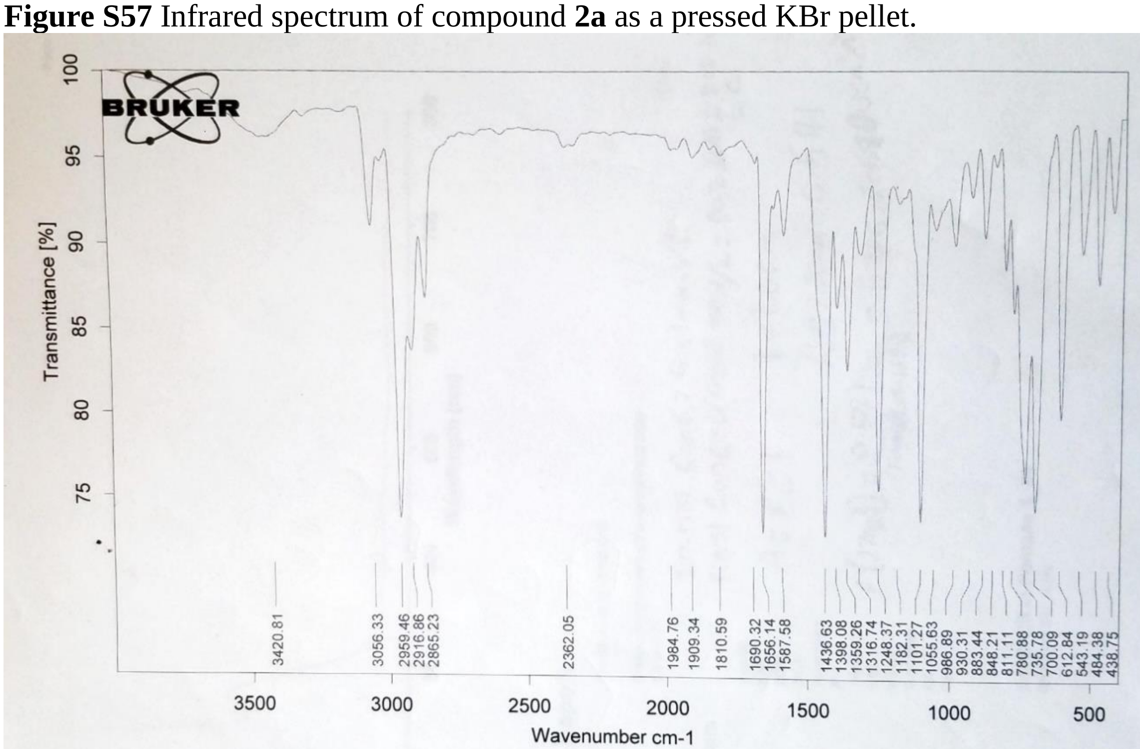


Figure S58 Infrared spectrum of compound **3** as a pressed KBr pellet.

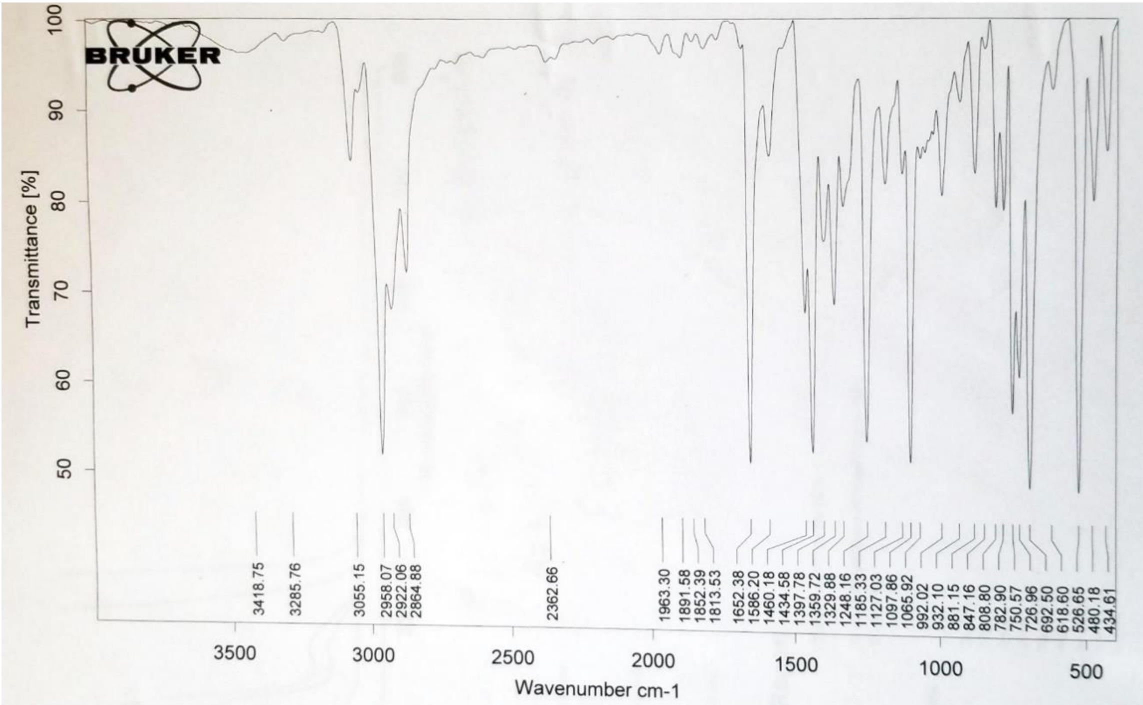


Figure S59 Infrared spectrum of compound **4** as a pressed KBr pellet.

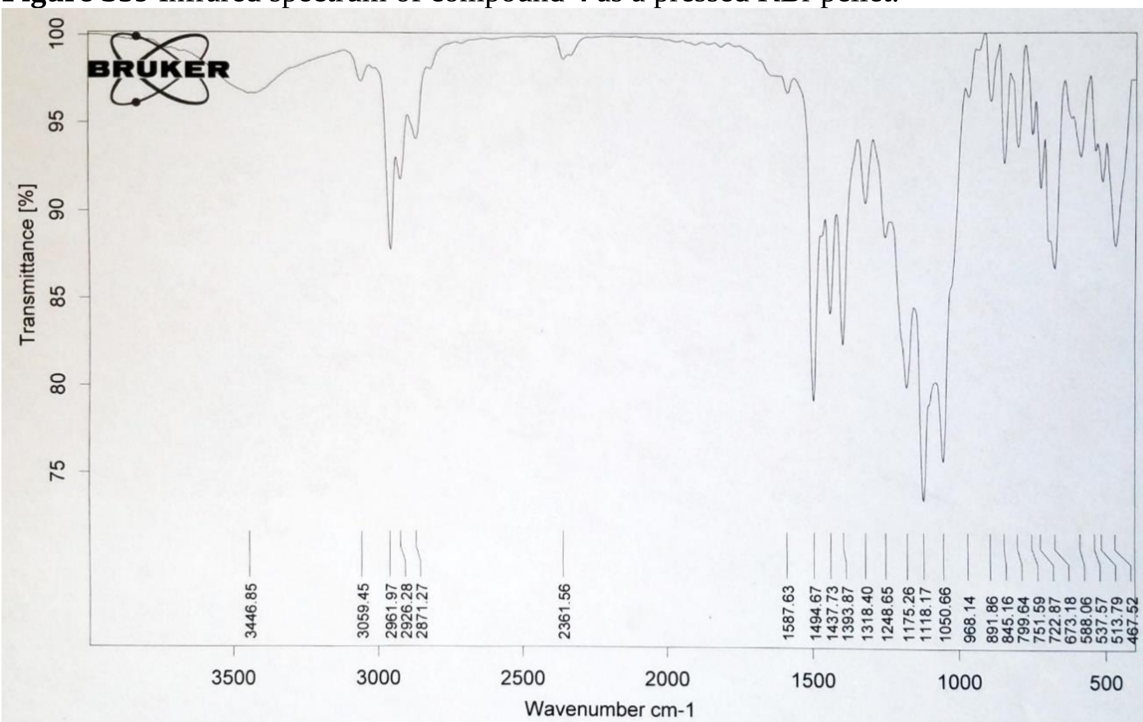


Figure S60 Infrared spectrum of compound **5** as a pressed KBr pellet.

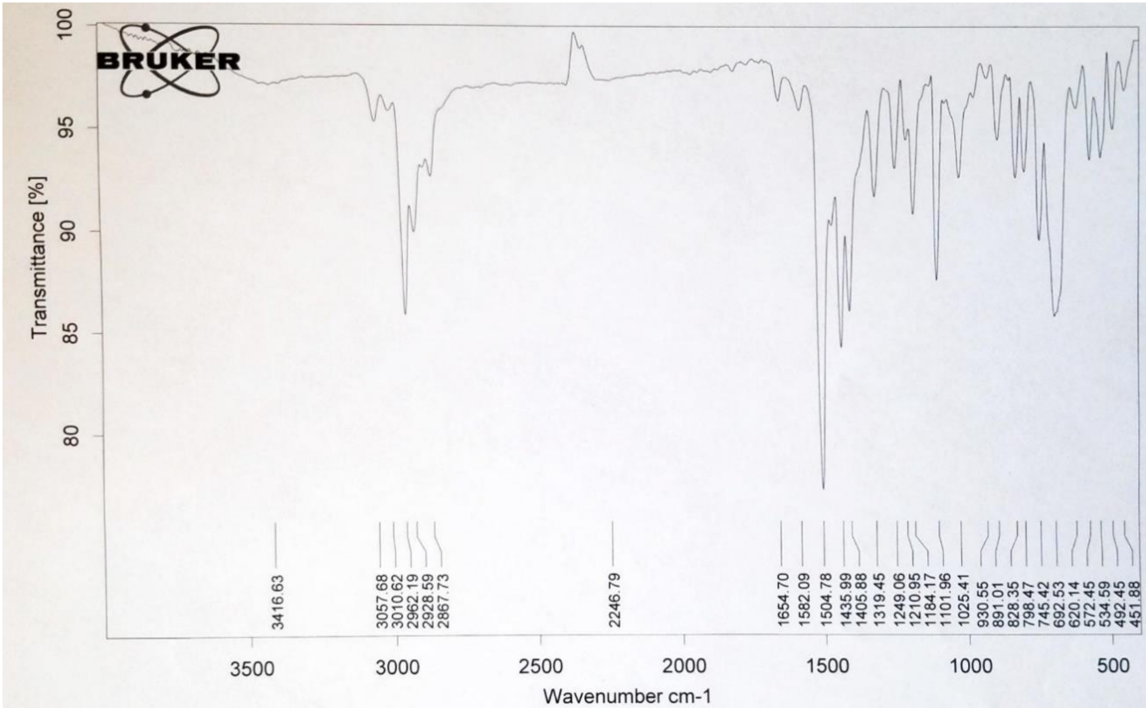


Figure S61 Infrared spectrum of compound **6** as a pressed KBr pellet.

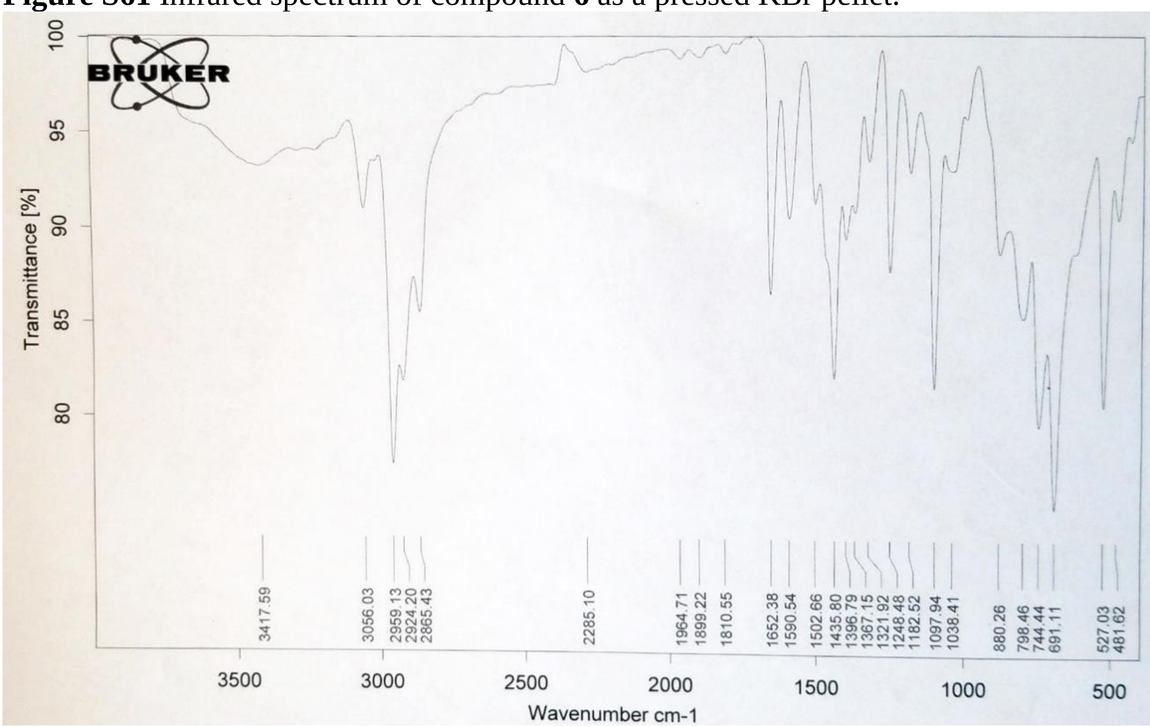


Figure S62 Infrared spectrum of compound **7** as a pressed KBr pellet.

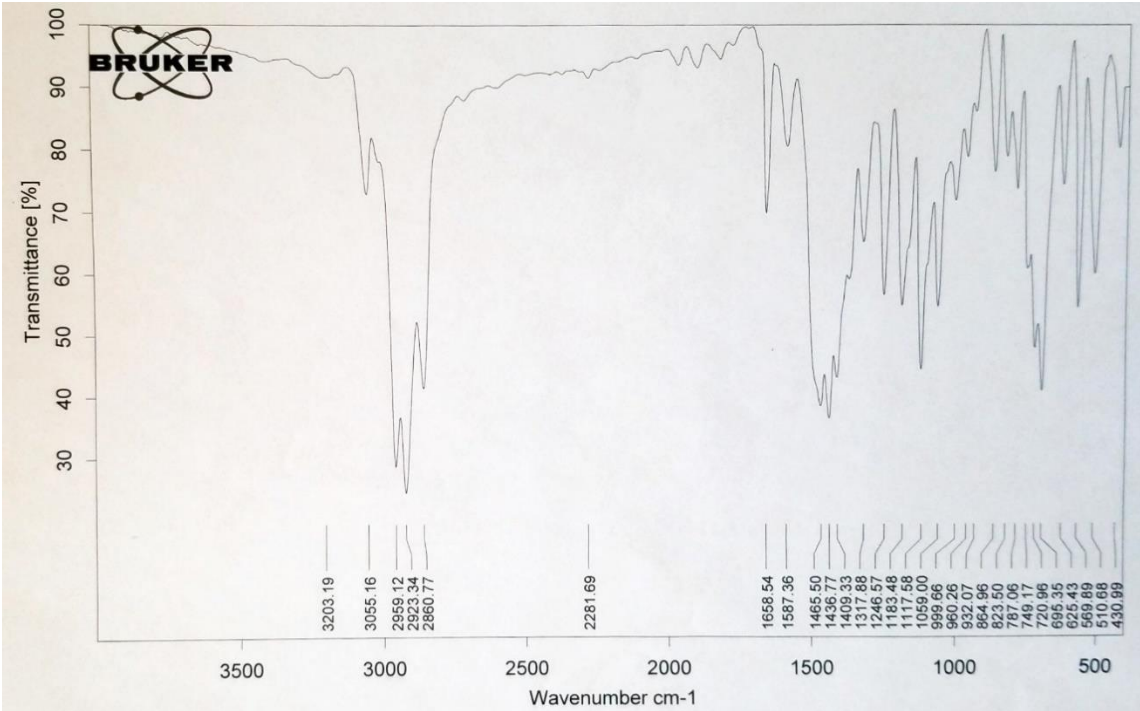


Figure S63 Infrared spectrum of compound **8a** as a pressed KBr pellet.

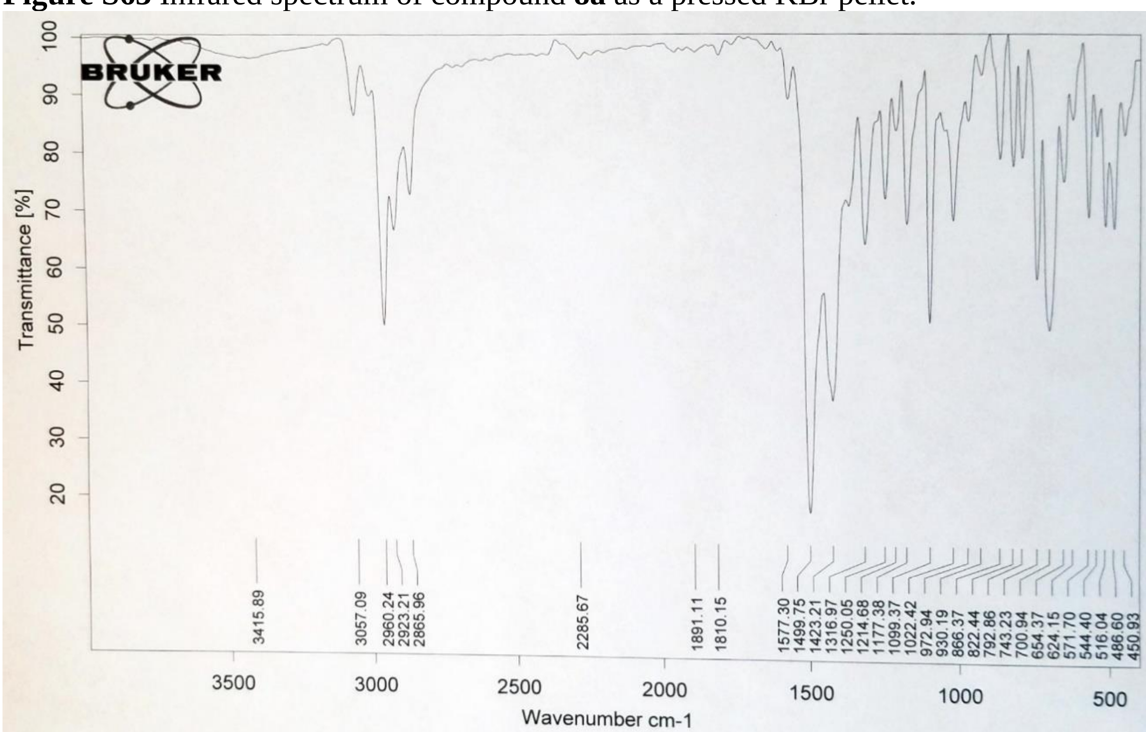


Figure S64 Infrared spectrum of compound **9** as a pressed KBr pellet.

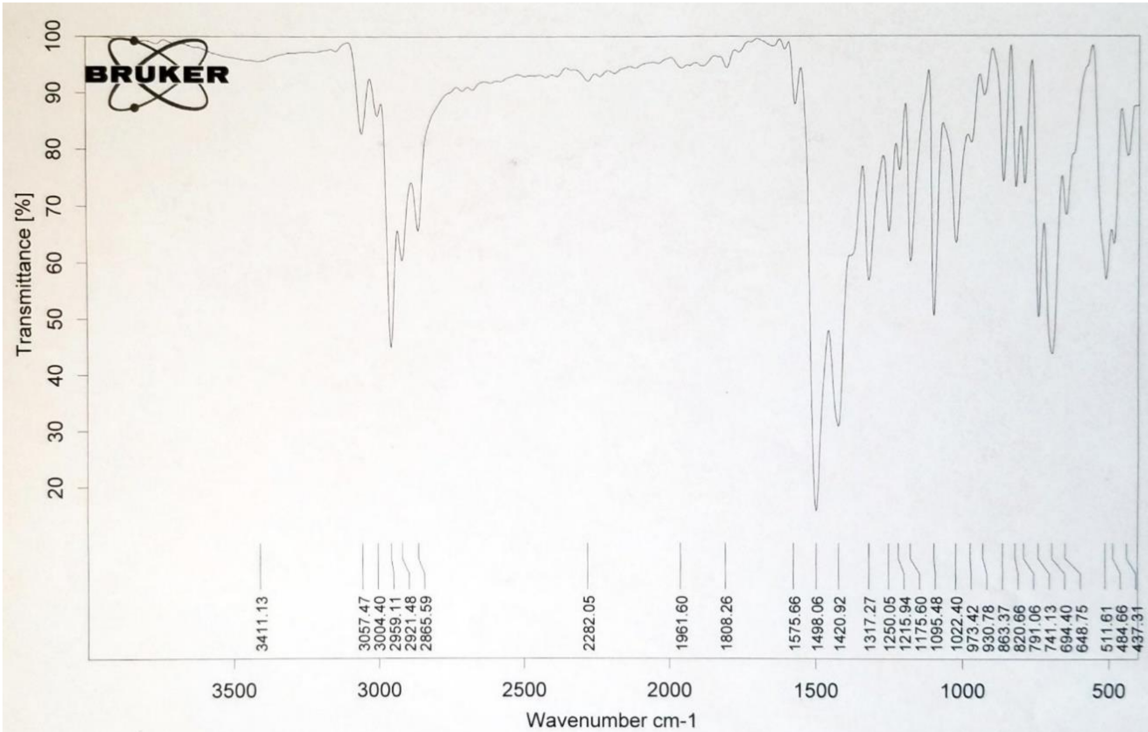


Figure S65 Infrared spectrum of compound **10** as a pressed KBr pellet.

4 - SC-XRD of Compounds 2-10

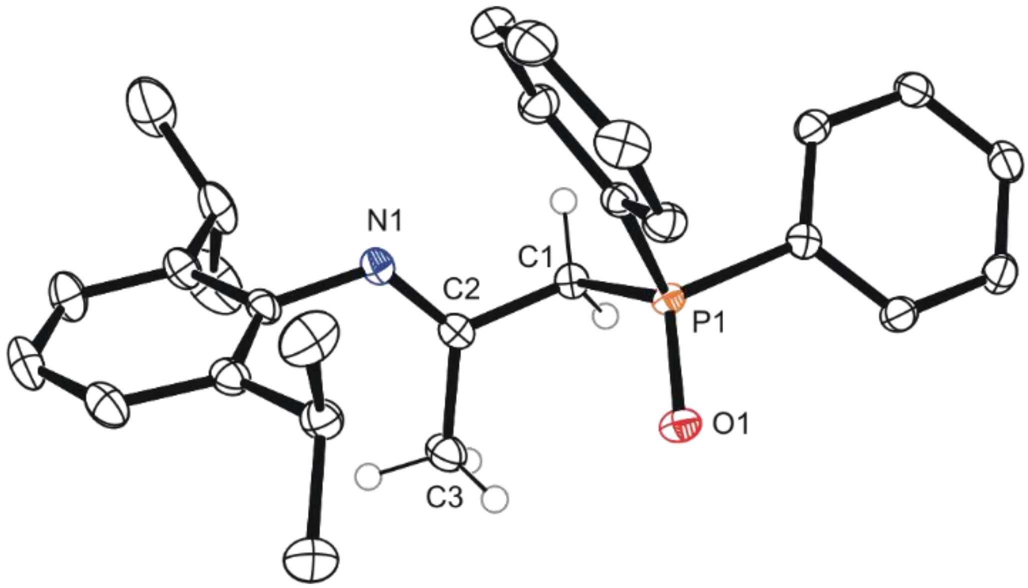


Figure S66 Single crystal structure of **2a** (imine isomer) in the solid state. Anisotropic displacement ellipsoids are set to 50 % probability and hydrogen atoms except for C1-H and C3-H are omitted for clarity.

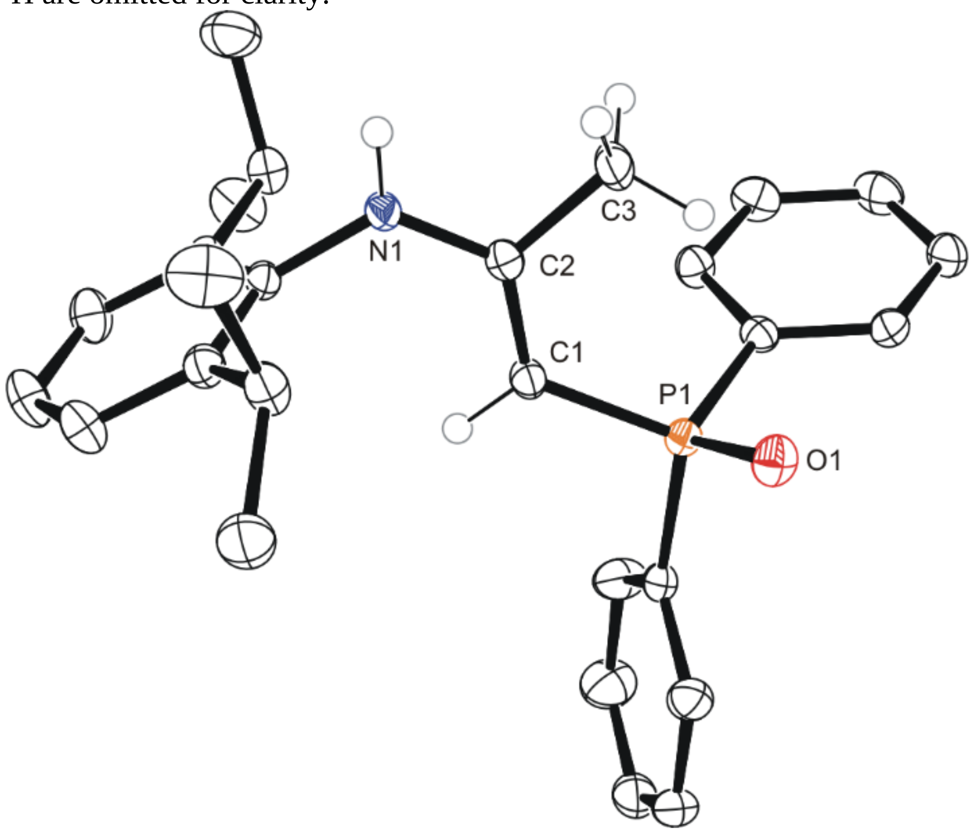


Figure S67 Single crystal structure of **2b** ((E)-enamine isomer) in the solid state. Anisotropic displacement ellipsoids are set to 50 % probability and hydrogen atoms except for C1-H and C3-H are omitted for clarity.

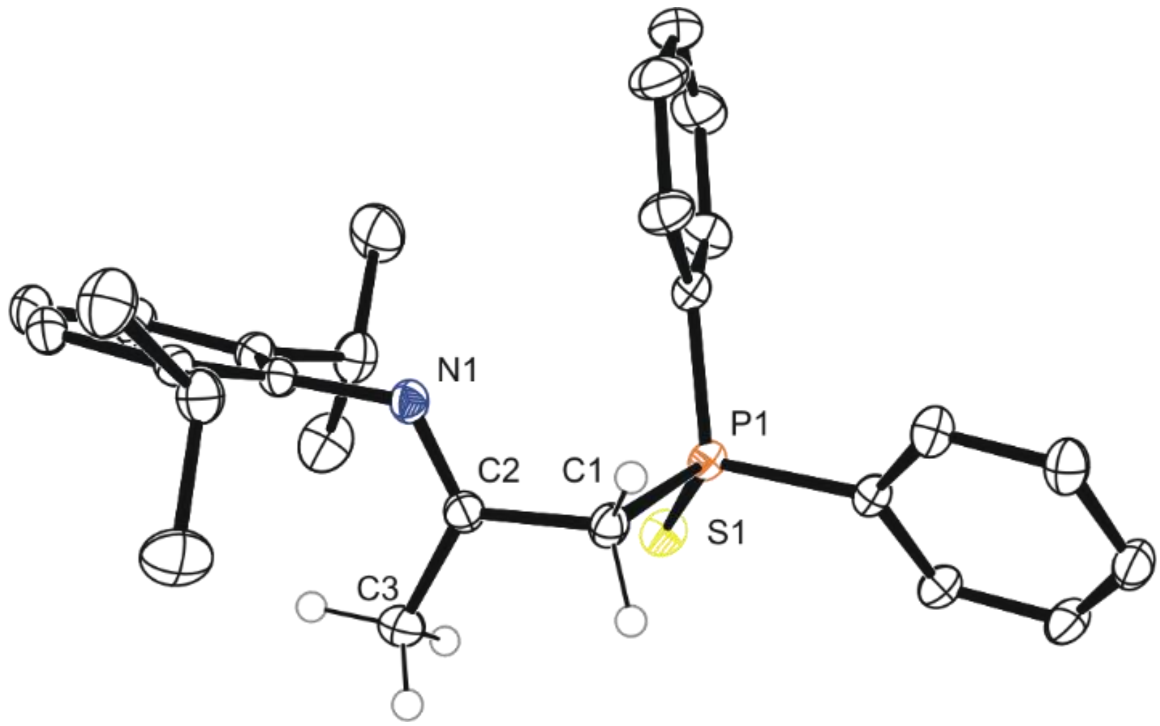


Figure S68 Single crystal structure of **3** in the solid state. Anisotropic displacement ellipsoids are set to 50 % probability and hydrogen atoms except for C1-H and C3-H are omitted for clarity.

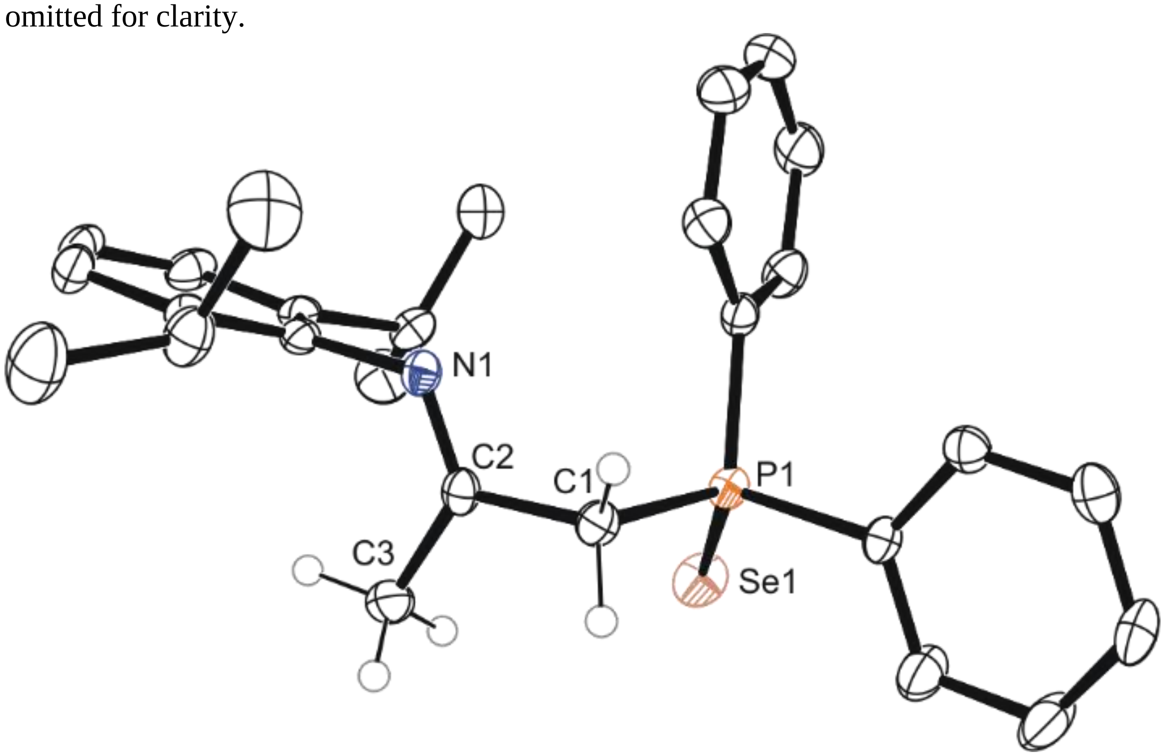


Figure S69 Single crystal structure of **4** in the solid state. Anisotropic displacement ellipsoids are set to 50 % probability and hydrogen atoms except for C1-H and C3-H are omitted for clarity.

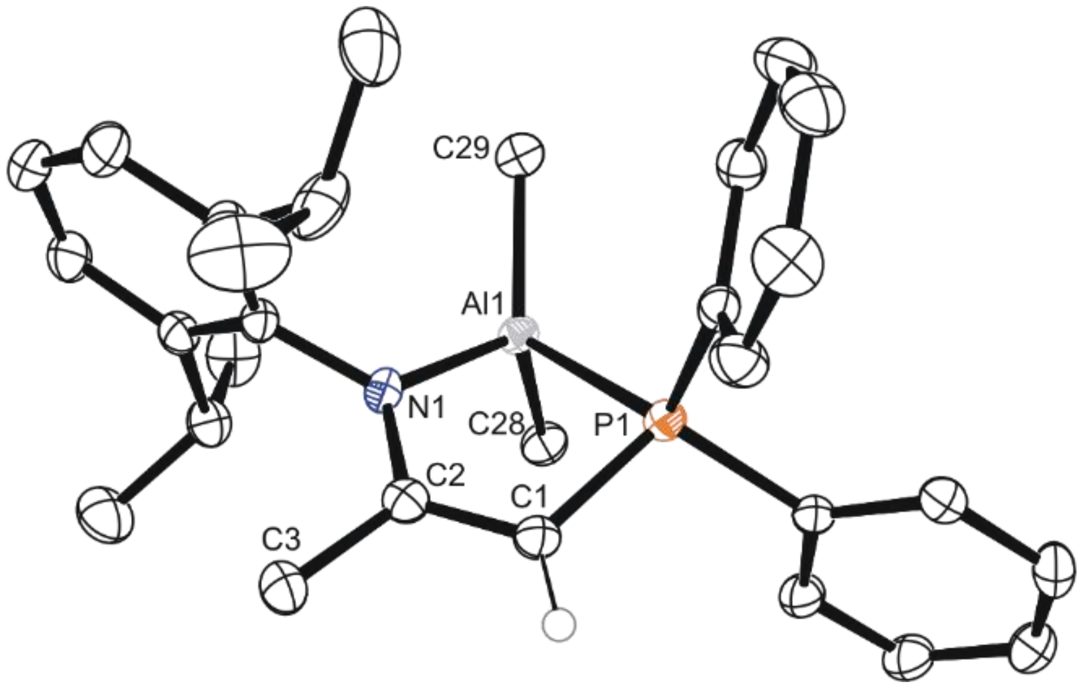


Figure S70 Single crystal structure of **1-AlMe2** in the solid state. Anisotropic displacement ellipsoids are set to 50 % probability and hydrogen atoms except for C1-H are omitted for clarity.

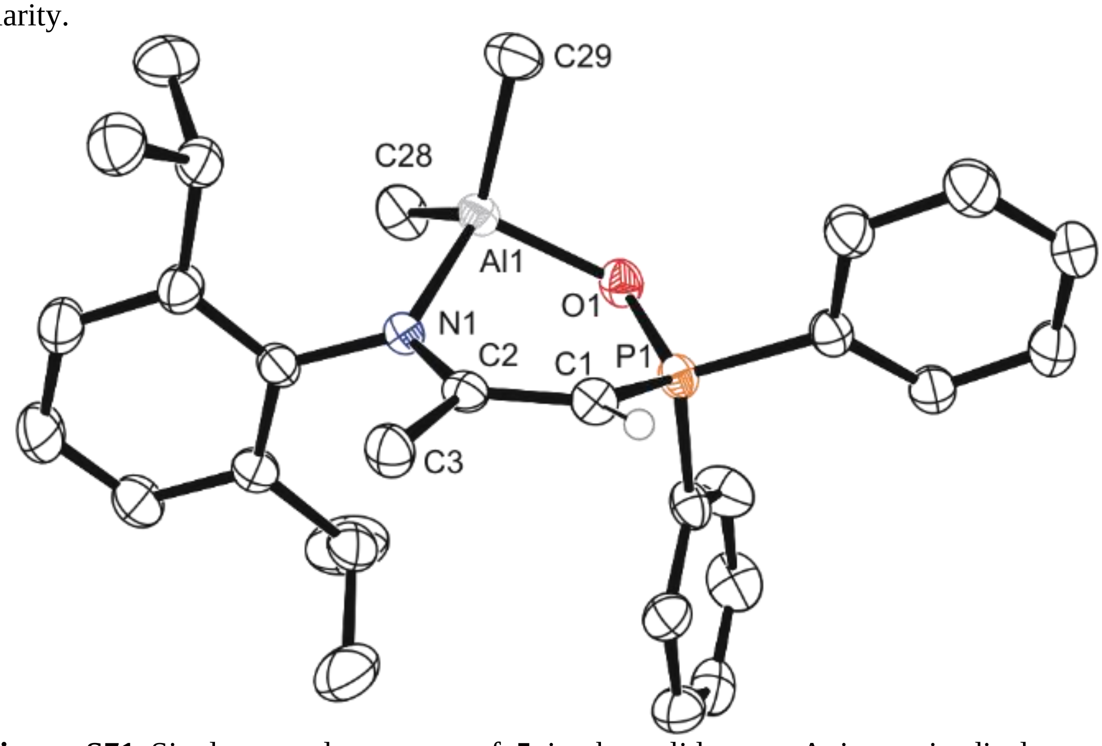


Figure S71 Single crystal structure of **5** in the solid state. Anisotropic displacement ellipsoids are set to 50 % probability and hydrogen atoms except for C1-H are omitted for clarity.

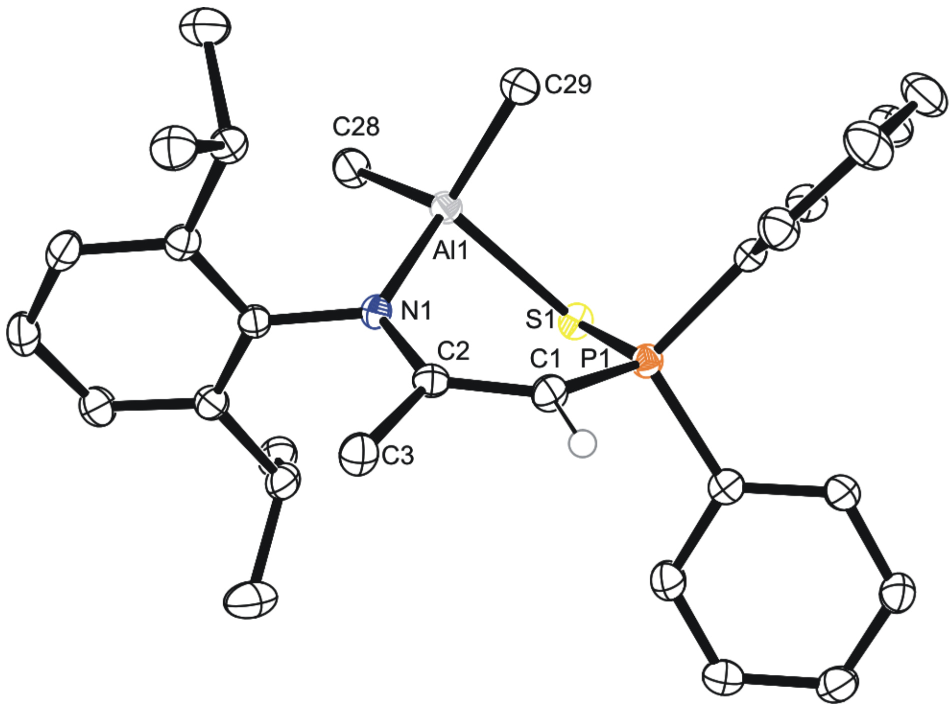


Figure S72 Single crystal structure of **6** in the solid state. Anisotropic displacement ellipsoids are set to 50 % probability and hydrogen atoms except for C1-H are omitted for clarity.

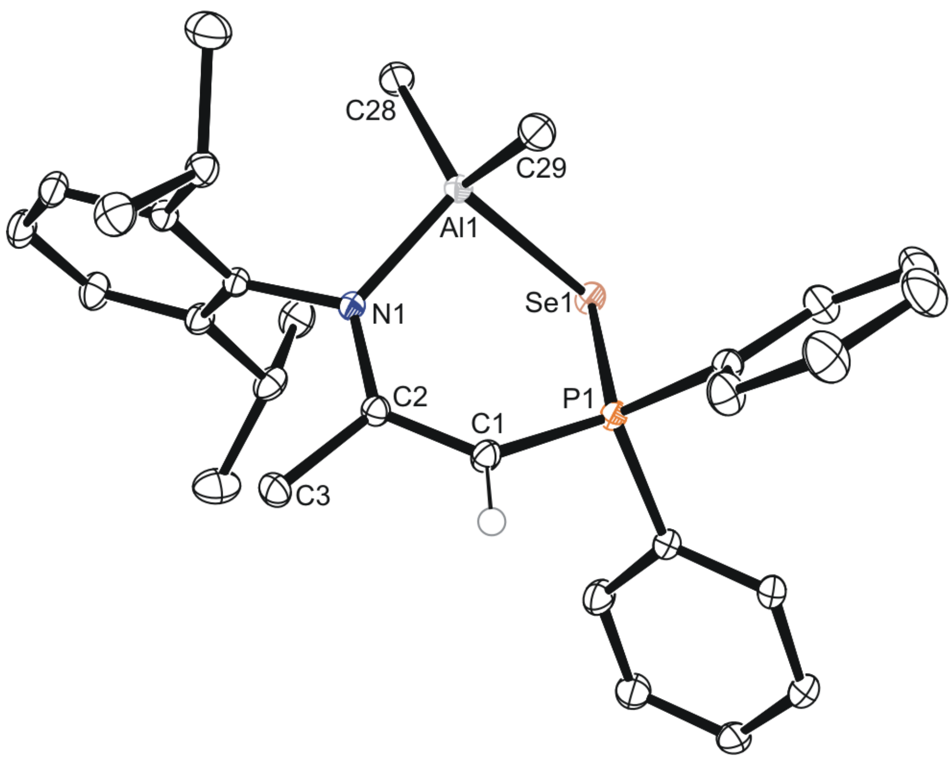


Figure S73 Single crystal structure of **7** in the solid state. Anisotropic displacement ellipsoids are set to 50 % probability and hydrogen atoms except for C1-H are omitted for clarity.

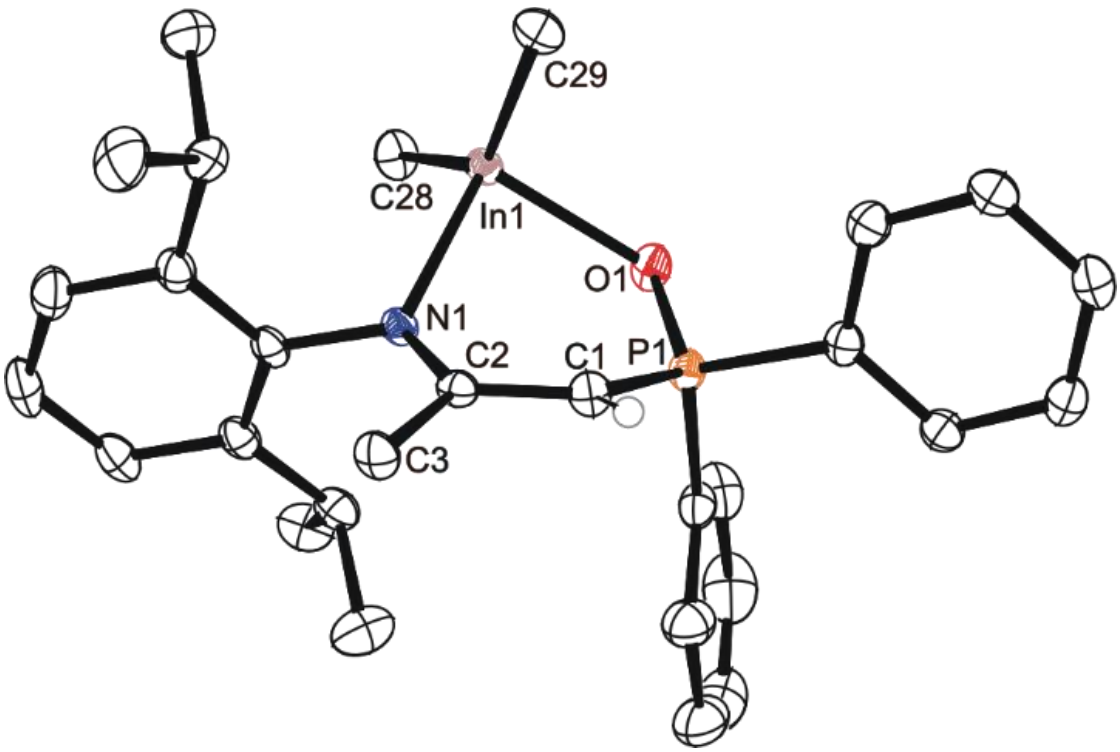


Figure S74 Single crystal structure of **8a** in the solid state. Anisotropic displacement ellipsoids are set to 50 % probability and hydrogen atoms except for C1-H are omitted for clarity.

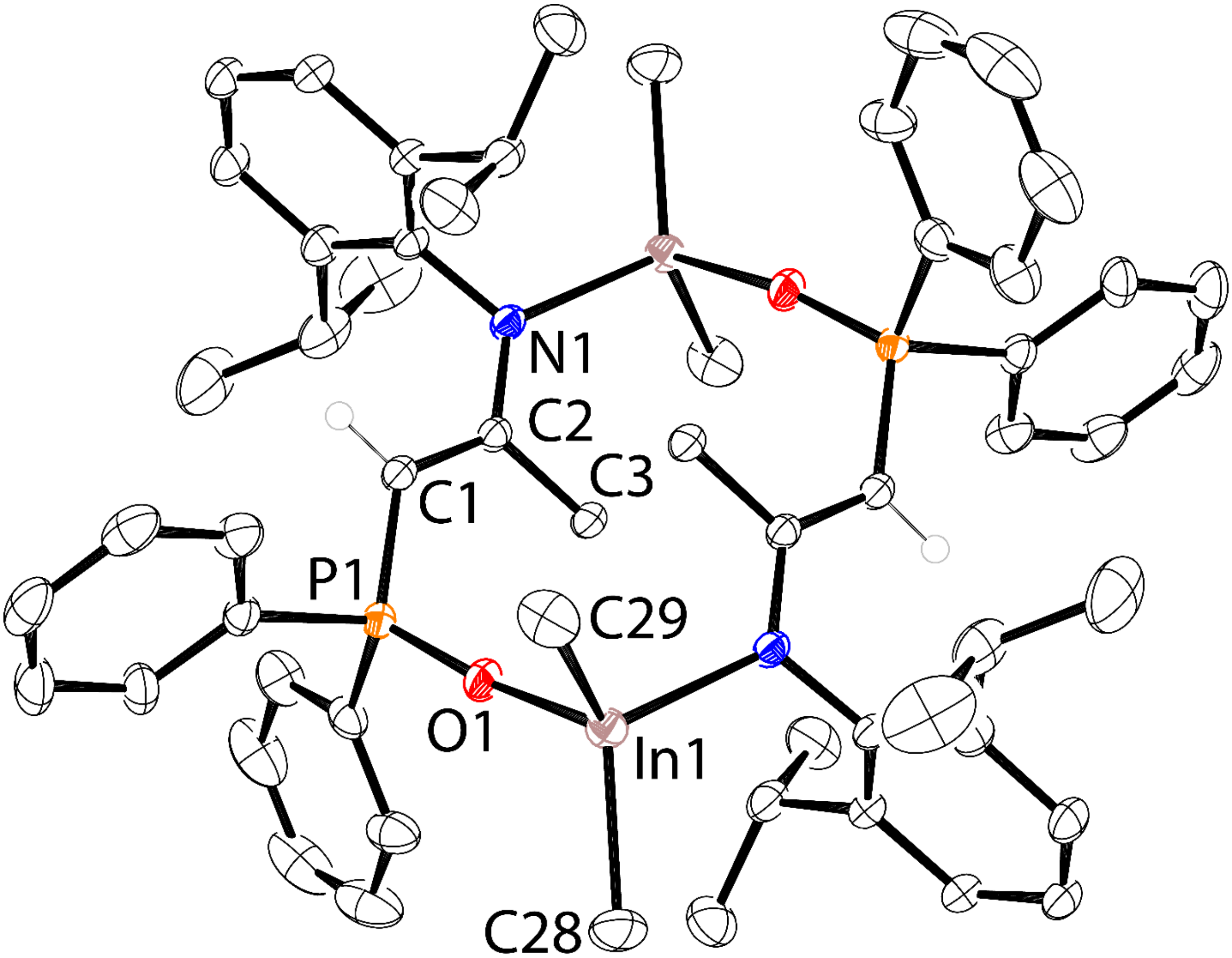


Figure S75 Single crystal structure of **8b** in the solid state. Anisotropic displacement ellipsoids are set to 50 % probability and hydrogen atoms except for C1-H are omitted for clarity.

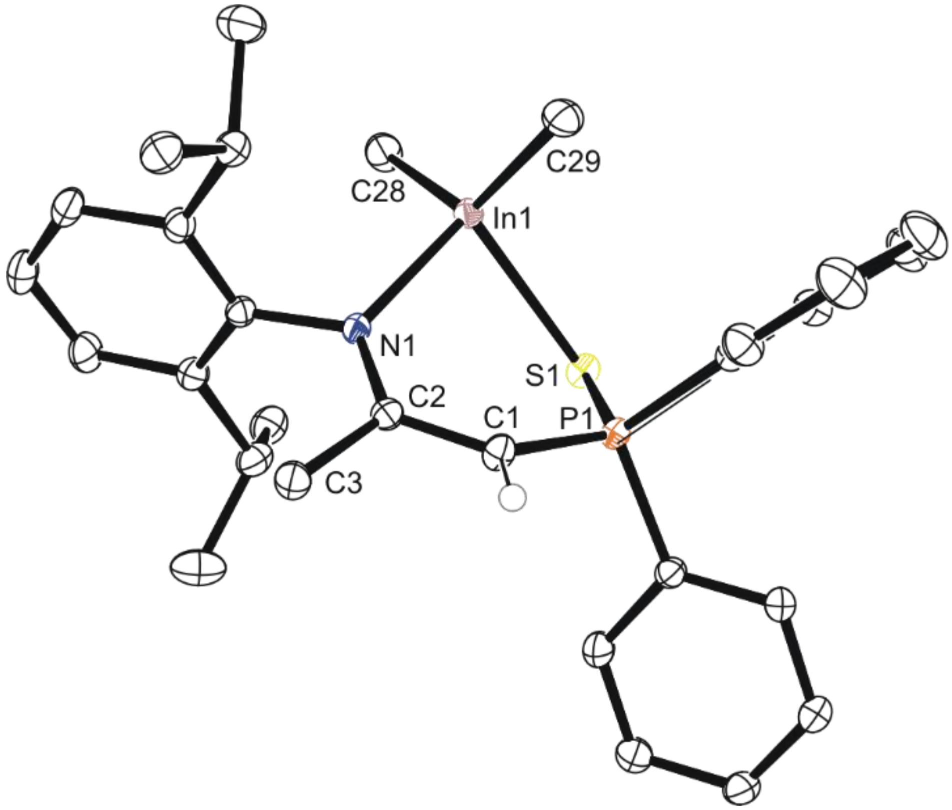


Figure S76 Single crystal structure of **9** in the solid state. Anisotropic displacement ellipsoids are set to 50 % probability and hydrogen atoms except for C1-H are omitted for clarity.

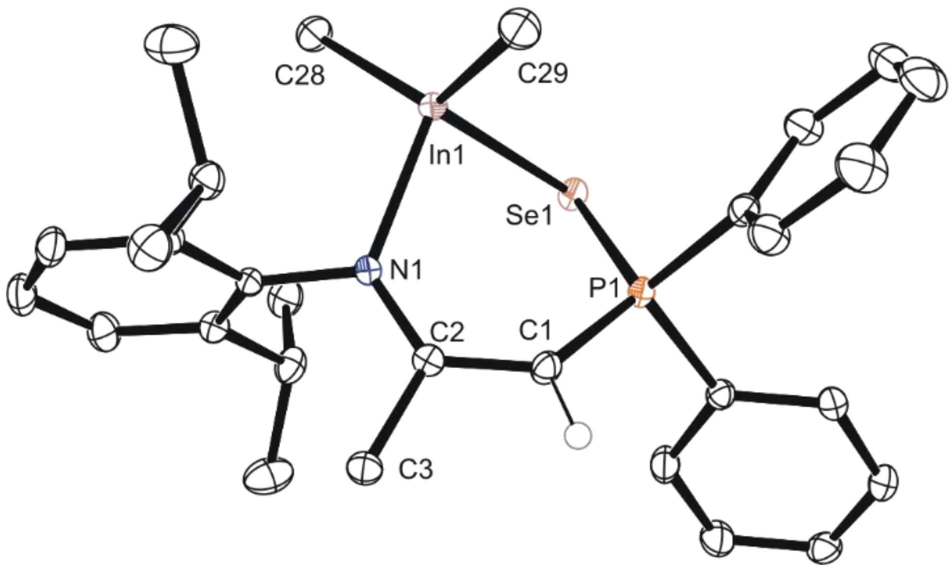


Figure S77 Single crystal structure of **10** in the solid state. Anisotropic displacement ellipsoids are set to 50 % probability and hydrogen atoms except for C1-H are omitted for clarity.

5 - Crystallography Table and Additional Refinement Details

Table S1 Data derived from single crystal X-ray diffraction of crystals of compounds **2-10** and the dimethyl aluminum chelate of compound **1**.

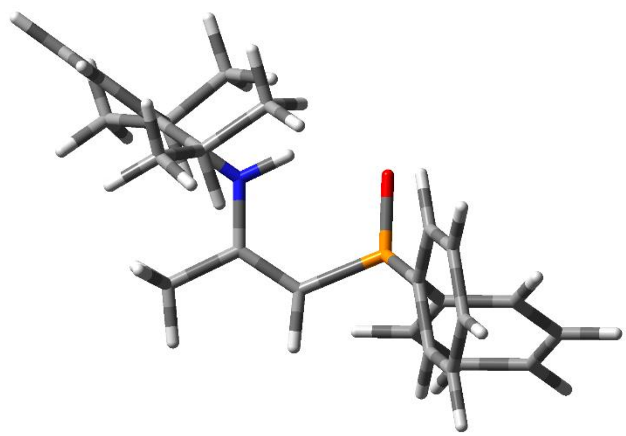
Compound	**2a**	**2b**	**3**	**4**	**5**	**6**	**7**	**8a**	**8b**	**9**	**10**
Empirical formula	$C_{27}H_{32}NOP$	$C_{27}H_{32}NOP$	$C_{27}H_{32}NPS$	$C_{27}H_{32}NPSe$	$C_{29}H_{37}AlNOP$	$C_{29}H_{37}AlNPS$	$C_{29}H_{37}AlNPSe$	$C_{29}H_{37}InNOP$	$C_{58}H_{74}In_2N_2O_2P_2$	$C_{29}H_{37}InNPS$	$C_{29}H_{37}InNPSe$
Formula weight	417.5	417.5	433.56	480.46	473.54	489.6	536.5	561.38	1122.77	577.44	624.34
Temperature/K	100	125	125	125	125	100	100	125	100	100	100
Crystal system	monoclinic	monoclinic	monoclinic	Monoclinic	monoclinic	orthorhombic	orthorhombic	monoclinic	Monoclinic	orthorhombic	orthorhombic
Space group	$P2_1/c$	$P2_1/n$	$P2_1/c$	$P2_1/c$	$P2_1/c$	Pbca	Pbca	$P2_1$	$P2_1/n$	Pbca	Pbca
a/Å	8.5833(3)	12.3073(3)	8.9711(4)	25.7547(14)	33.496(2)	15.5808(12)	15.4976(3)	8.6720(5)	13.1361(4)	15.7440(7)	15.6585(2)
b/Å	25.7233(8)	9.7254(3)	23.6926(10)	21.8166(12)	10.7213(7)	16.7471(13)	16.8142(4)	17.5977(8)	14.1548(5)	16.8069(6)	16.9162(2)
c/Å	11.4020(3)	20.0034(4)	12.4579(6)	17.7353(9)	15.6598(11)	21.2634(18)	21.3983(4)	18.1245(10)	15.2502(6)	21.3702(10)	21.4668(4)
α/°	90	90	90	90	90	90	90	90	90	90	90
β/°	111.2060(10)	90.1310(10)	110.367(2)	100.0610(10)	103.480(2)	90	90	95.677(2)	99.9240(10)	90	90
γ/°	90	90	90	90	90	90	90	90	90	90	90
Volume/$Å^3$	2346.99(13)	2394.27(11)	2482.4(2)	9811.9(9)	5468.8(7)	5548.3(8)	5576.0(2)	2752.4(3)	2793.18(17)	5654.7(4)	5686.18(14)
$\rho_{calc}g/cm^3$	1.182	1.158	1.16	1.301	1.15	1.172	1.278	1.355	1.335	1.357	1.459
μ/mm^{-1}	0.135	0.132	0.208	1.609	0.153	0.223	1.453	0.937	0.923	0.983	2.184
F(000)	896	896	928	4000	2032	2096	2240	1160	1160	2384	2528
Crystal size/mm^3	0.1 × 0.1 × 0.02	0.2 × 0.08 × 0.05	0.32 × 0.15 × 0.15	0.1 × 0.1 × 0.1	0.3 × 0.2 × 0.05	0.14 × 0.1 × 0.09	0.19 × 0.12 × 0.09	0.44 × 0.24 × 0.24	0.307 × 0.225 × 0.22	0.14 × 0.13 × 0.1	0.293 × 0.218 × 0.183
2Θ range for data collection/°	4.146 to 50.692	3.882 to 66.292	3.888 to 66.312	2.462 to 58.252	4.548 to 50.75	4.052 to 56.954	4.626 to 75.568	4.63 to 54.166	4.754 to 67.496	4.608 to 64.06	5.176 to 56.572
Reflections collected	22736	47727	128850	123334	361528	241435	223922	95735	439311	176943	48155
Data/restraints/parameters	4290/0/276	8813/0/293	9447/0/276	25247/0/1101	10011/0/623	6970/0/309	14895/0/305	12036/1/609	11172/0/305	9844/0/305	6914/0/309
Goodness-of-fit on F^2	1.081	1.029	1.041	1.002	1.112	1.123	1.033	1.071	1.112	1.133	1.019
Final R indexes [I>=2σ (I)]	R_1 = 0.0356, wR_2 = 0.0870	R_1 = 0.0488, wR_2 = 0.1021	R_1 = 0.0321, wR_2 = 0.0903	R_1 = 0.0413, wR_2 = 0.0787	R_1 = 0.0413, wR_2 = 0.1025	R_1 = 0.0383, wR_2 = 0.0819	R_1 = 0.0289, wR_2 = 0.0670	R_1 = 0.0158, wR_2 = 0.0416	R_1 = 0.0239, wR_2 = 0.0652	R_1 = 0.0255, wR_2 = 0.0528	R_1 = 0.0150, wR_2 = 0.0375
Final R indexes [all data]	R_1 = 0.0384, wR_2 = 0.0888	R_1 = 0.0820, wR_2 = 0.1190	R_1 = 0.0354, wR_2 = 0.0930	R_1 = 0.0899, wR_2 = 0.0921	R_1 = 0.0509, wR_2 = 0.1111	R_1 = 0.0614, wR_2 = 0.0982	R_1 = 0.0398, wR_2 = 0.0710	R_1 = 0.0160, wR_2 = 0.0417	R_1 = 0.0261, wR_2 = 0.0677	R_1 = 0.0392, wR_2 = 0.0623	R_1 = 0.0165, wR_2 = 0.0383

Compound **2b** is the (E)-enamine isomer of **2**. In the crystal structure hydrogen bonding between the donor N1-H and acceptor O1 across the symmetry element 0.5-X,0.5+Y,1.5-Z and 0.5-X,-0.5+Y,1.5-Z is observed forming a chain structure. The C3 methyl group win this structure is modelled with two component rotational disorder about the C3-C2 bond in a 59:41 ratio.

6 - Computational Analysis of the Isomers of Compound 2

Calculations were performed using Gaussian 16 (Revision B.01)[1] using the M06-2X[2] functional and def2-TZVP[3] basis set with the iefpcm solvent model (chloroform). Initial structures were optimized at the B3LYP/6-31G(d)[4,5] level with the GD3BJ dispersion correction[6] as a starting point for the higher-level calculations. All isomers and conformers are local minima from frequency calculations (no imaginary frequencies).

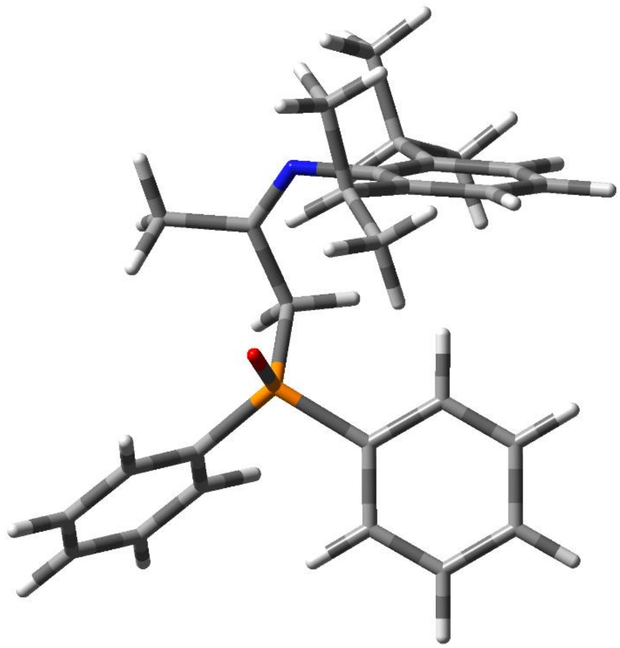
(Z)-enamine, Energy: -1518.983201 Hartrees



P	1.685300	-0.217200	-0.221100
O	0.920200	-0.275600	-1.508700
N	-1.419500	-0.357800	0.039100
C	0.670700	-0.374800	1.211200
H	1.162800	-0.404100	2.174500
C	-0.690700	-0.356500	1.185200
C	-1.479300	-0.344100	2.462700
H	-2.031100	0.594200	2.554800
H	-0.820000	-0.452000	3.319100
H	-2.215800	-1.149900	2.465300
C	-2.809200	-0.038300	-0.021300
C	-3.741900	-1.078500	-0.112500
C	-5.092200	-0.745200	-0.185700
H	-5.837400	-1.526500	-0.253000
C	-5.496700	0.580000	-0.165300
H	-6.550500	0.822500	-0.220300
C	-4.559700	1.597400	-0.068000
H	-4.894700	2.625700	-0.045300
C	-3.200000	1.307900	0.005800
C	-3.271600	-2.518100	-0.170200
H	-2.395100	-2.599500	0.476900
C	-2.825700	-2.864900	-1.594700
H	-2.029300	-2.203300	-1.936300
H	-3.668200	-2.769900	-2.283600
H	-2.459800	-3.892000	-1.641000
C	-4.312800	-3.521000	0.317600
H	-4.699500	-3.253400	1.302200
H	-3.864200	-4.512800	0.383600

H	-5.155700	-3.591000	-0.372600
C	-2.154600	2.406100	0.070100
H	-1.347900	2.063800	0.724000
C	-1.548600	2.641900	-1.318400
H	-1.068900	1.743400	-1.709600
H	-0.797100	3.432500	-1.271900
H	-2.327700	2.950500	-2.019100
C	-2.684800	3.717900	0.641900
H	-3.389000	4.196300	-0.041500
H	-1.857200	4.411300	0.794800
H	-3.186800	3.566700	1.598900
C	2.986400	-1.471900	-0.097800
C	2.792500	-2.635200	0.641800
H	1.873800	-2.767700	1.200300
C	3.770700	-3.621300	0.663600
H	3.615900	-4.522200	1.242900
C	4.944500	-3.449700	-0.056700
H	5.707500	-4.217400	-0.038500
C	5.142900	-2.292300	-0.801200
H	6.057800	-2.158700	-1.363600
C	4.168600	-1.306600	-0.820400
H	4.327000	-0.402200	-1.396700
C	2.574800	1.355600	-0.077600
C	3.493300	1.587000	0.945400
H	3.729400	0.799200	1.652500
C	4.114900	2.821000	1.056200
H	4.828900	2.997500	1.850200
C	3.823100	3.830100	0.144100
H	4.310800	4.792600	0.230400
C	2.912300	3.602700	-0.877900
H	2.689400	4.386200	-1.590700
C	2.287700	2.365900	-0.989400
H	1.577600	2.171000	-1.783700
H	-0.877400	-0.336500	-0.821400

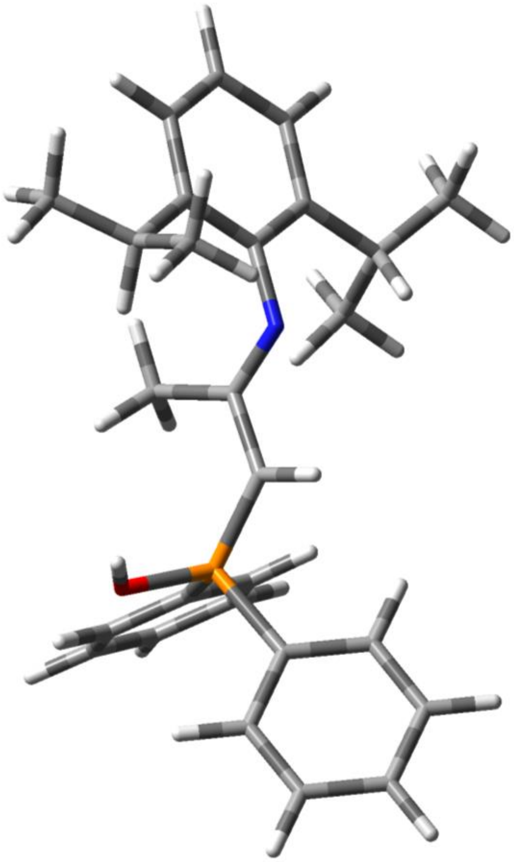
(Z)-imine 2, Energy: -1518.978868 Hartrees



P	1.463200	-0.024400	0.415400
O	1.096900	-0.164400	1.849200
N	-1.673600	-1.829500	0.393000
C	0.340300	-0.945700	-0.709700
C	-0.466900	-1.983100	0.038400
C	0.254400	-3.253000	0.369300
H	1.144000	-3.021700	0.962500
H	0.594000	-3.745400	-0.544500
H	-0.391200	-3.922400	0.931500
C	-2.360200	-0.617700	0.144300
C	-2.557300	0.261700	1.221600
C	-3.243800	1.449500	0.984500
H	-3.393000	2.148900	1.796600
C	-3.729200	1.758400	-0.276000
H	-4.251800	2.691400	-0.444200
C	-3.552600	0.863500	-1.321100
H	-3.946300	1.109400	-2.298100
C	-2.878400	-0.340000	-1.133700
C	-2.073900	-0.113300	2.608400
H	-1.145700	-0.670400	2.491800
C	-3.107000	-1.024900	3.279000
H	-3.290300	-1.914800	2.675900
H	-4.053600	-0.494100	3.407700
H	-2.756100	-1.340600	4.263500
C	-1.762800	1.091800	3.489200
H	-1.078600	1.778200	2.987700
H	-1.288400	0.756400	4.412400
H	-2.666300	1.639700	3.766100
C	-2.750200	-1.360700	-2.250000
H	-1.742000	-1.779900	-2.215100
C	-3.733000	-2.512000	-2.008400

H	-3.568000	-2.969700	-1.032800
H	-3.621100	-3.279500	-2.776400
H	-4.758600	-2.138100	-2.044400
C	-2.948400	-0.777500	-3.645300
H	-3.980700	-0.458700	-3.801700
H	-2.724800	-1.537000	-4.395300
H	-2.296200	0.079900	-3.821000
C	1.410400	1.679600	-0.190000
C	2.570100	2.430900	-0.365100
H	3.541200	1.978300	-0.207600
C	2.486600	3.765300	-0.741400
H	3.391600	4.342600	-0.878300
C	1.246400	4.355600	-0.939100
H	1.183200	5.395200	-1.233500
C	0.085100	3.613600	-0.757100
H	-0.884500	4.071300	-0.905000
C	0.164300	2.281000	-0.384400
H	-0.751900	1.717900	-0.234800
C	3.123100	-0.648900	0.072900
C	3.655500	-0.666200	-1.216300
H	3.079700	-0.281400	-2.051300
C	4.929800	-1.164800	-1.433800
H	5.342600	-1.175500	-2.434000
C	5.676400	-1.650800	-0.364900
H	6.671100	-2.041500	-0.536700
C	5.149900	-1.635700	0.918400
H	5.731500	-2.014200	1.748700
C	3.872800	-1.133600	1.139400
H	3.446200	-1.115000	2.134500
H	-0.309700	-0.227900	-1.205000
H	0.955700	-1.431700	-1.468800

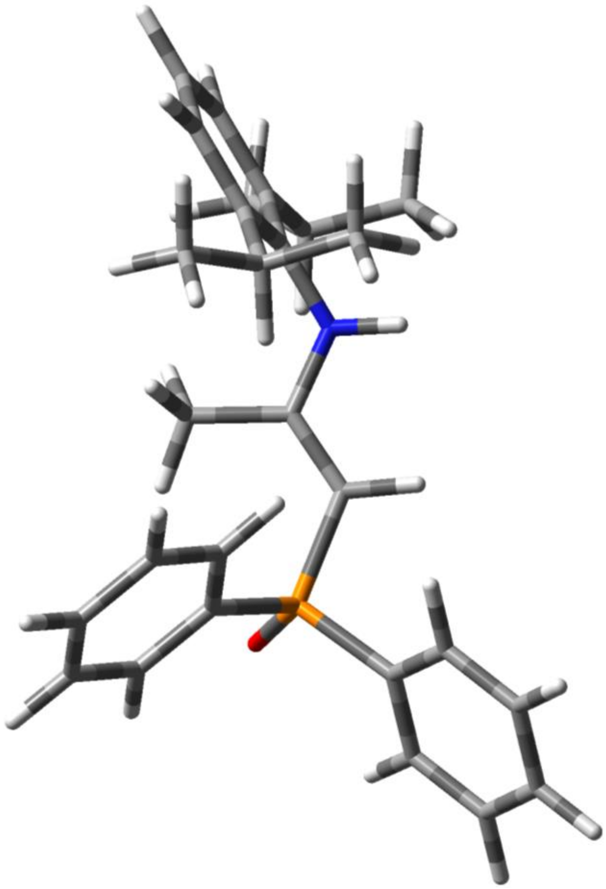
(E)-ylide, Energy: -1518.942231 Hartrees



P	2.057300	-0.636100	-0.357800
O	2.311100	-1.622900	-1.616000
N	-1.707200	-0.522300	0.779800
C	0.608400	-0.748300	0.472600
C	-0.694200	-0.358400	0.003000
C	-0.805000	0.237100	-1.387100
H	-0.303200	-0.399100	-2.119500
H	-0.316000	1.212400	-1.421000
H	-1.843600	0.363700	-1.684100
C	-3.019100	-0.210800	0.400100
C	-3.639100	0.916400	0.971600
C	-4.967000	1.186900	0.656100
H	-5.454400	2.051600	1.088200
C	-5.681100	0.364200	-0.200600
H	-6.712600	0.589500	-0.439500
C	-5.069600	-0.757700	-0.741500
H	-5.639000	-1.399400	-1.401300
C	-3.745300	-1.070000	-0.451200
C	-2.841200	1.812900	1.897100
H	-2.177400	1.157300	2.466600
C	-3.702800	2.596200	2.883800
H	-4.391300	1.943300	3.422200
H	-4.289300	3.367200	2.379500
H	-3.065700	3.098400	3.613200

C	-1.959100	2.776000	1.096000
H	-1.276200	2.236200	0.440300
H	-1.366000	3.402400	1.766100
H	-2.580000	3.429900	0.478500
C	-3.095300	-2.335700	-0.980300
H	-2.075600	-2.088100	-1.284700
C	-2.998200	-3.378200	0.138900
H	-2.451400	-2.979800	0.993400
H	-2.488300	-4.276700	-0.215000
H	-4.000000	-3.662300	0.469500
C	-3.799100	-2.931300	-2.195500
H	-4.784000	-3.323600	-1.934100
H	-3.212400	-3.762100	-2.590100
H	-3.926400	-2.194300	-2.990300
C	3.410700	-0.968400	0.783100
C	3.330900	-0.576600	2.119100
H	2.434600	-0.095700	2.489100
C	4.400300	-0.803700	2.972700
H	4.332200	-0.502900	4.009800
C	5.553900	-1.414200	2.497600
H	6.386800	-1.590200	3.165900
C	5.640200	-1.797800	1.165900
H	6.538400	-2.272800	0.794100
C	4.572800	-1.574700	0.308100
H	4.639100	-1.875300	-0.729600
C	2.392100	0.938700	-1.173500
C	2.167200	2.103100	-0.439800
H	1.775000	2.034700	0.568700
C	2.422300	3.342000	-1.005900
H	2.241200	4.243100	-0.435000
C	2.902500	3.423400	-2.307800
H	3.097000	4.390900	-2.752200
C	3.129400	2.266200	-3.039700
H	3.500200	2.329700	-4.054300
C	2.877800	1.022100	-2.475500
H	3.048600	0.120900	-3.047800
H	0.665200	-1.221900	1.443400
H	1.943800	-2.502600	-1.456300

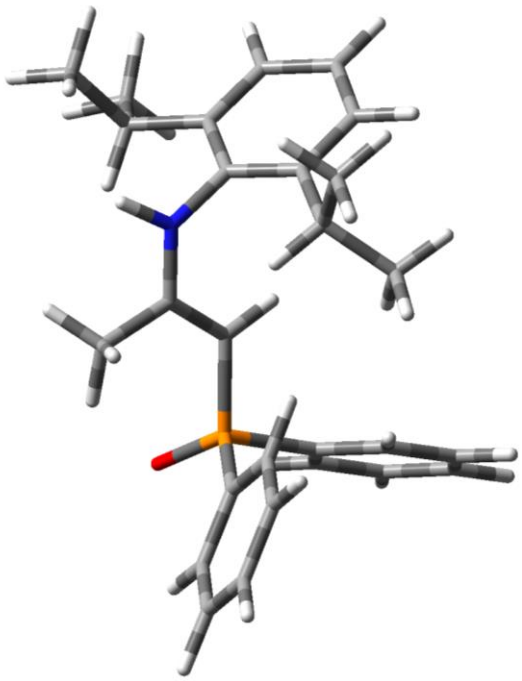
(E)-enamine 1, Energy: -1518.977889 Hartrees



P	-2.131000	-0.717000	-0.777000
O	-2.254200	-1.395100	-2.099500
N	1.670000	-0.818100	0.555800
C	-0.664300	-0.983500	0.170000
H	-0.776700	-1.524200	1.103300
C	0.570700	-0.569400	-0.211200
C	0.836600	0.176700	-1.486300
H	1.083800	1.219300	-1.269400
H	-0.021400	0.148500	-2.151400
H	1.693400	-0.260100	-2.001500
C	2.951800	-0.217400	0.350300
C	3.955100	-0.963500	-0.281200
C	5.199800	-0.366500	-0.461100
H	5.994900	-0.915400	-0.947700
C	5.431600	0.932300	-0.036300
H	6.404500	1.382700	-0.187600
C	4.421300	1.661700	0.570100
H	4.615600	2.678500	0.884500
C	3.163100	1.101900	0.773200
C	3.685600	-2.387300	-0.724700
H	2.641500	-2.436500	-1.041900
C	3.857600	-3.346400	0.457700
H	3.198700	-3.083800	1.285900

H	4.887500	-3.312900	0.820200
H	3.633700	-4.370400	0.155000
C	4.553800	-2.834000	-1.897300
H	4.507100	-2.124300	-2.724500
H	4.210200	-3.803800	-2.258600
H	5.598700	-2.947700	-1.602400
C	2.063500	1.872700	1.477600
H	1.105900	1.527700	1.082000
C	2.084700	1.555300	2.976400
H	1.979900	0.485000	3.159800
H	1.272900	2.073100	3.490100
H	3.030500	1.881000	3.414700
C	2.133500	3.380200	1.249500
H	2.996900	3.823700	1.748200
H	1.241600	3.855100	1.660400
H	2.190400	3.621400	0.186800
C	-2.321600	1.082000	-0.978700
C	-1.868800	1.995200	-0.028100
H	-1.361200	1.640400	0.861500
C	-2.050100	3.357200	-0.223200
H	-1.693200	4.061200	0.517600
C	-2.682900	3.816100	-1.372300
H	-2.819900	4.878600	-1.526500
C	-3.133300	2.911500	-2.324700
H	-3.621200	3.267500	-3.222900
C	-2.953500	1.548400	-2.128800
H	-3.288800	0.834900	-2.871200
C	-3.465700	-1.207000	0.347600
C	-3.530400	-0.760100	1.666700
H	-2.757900	-0.108500	2.059300
C	-4.583200	-1.145600	2.482200
H	-4.629900	-0.796600	3.505700
C	-5.578100	-1.980100	1.984200
H	-6.399300	-2.281000	2.621900
C	-5.518100	-2.427100	0.671900
H	-6.291300	-3.077900	0.284300
C	-4.463800	-2.040300	-0.146600
H	-4.400700	-2.383000	-1.171800
H	1.512900	-1.288400	1.435100

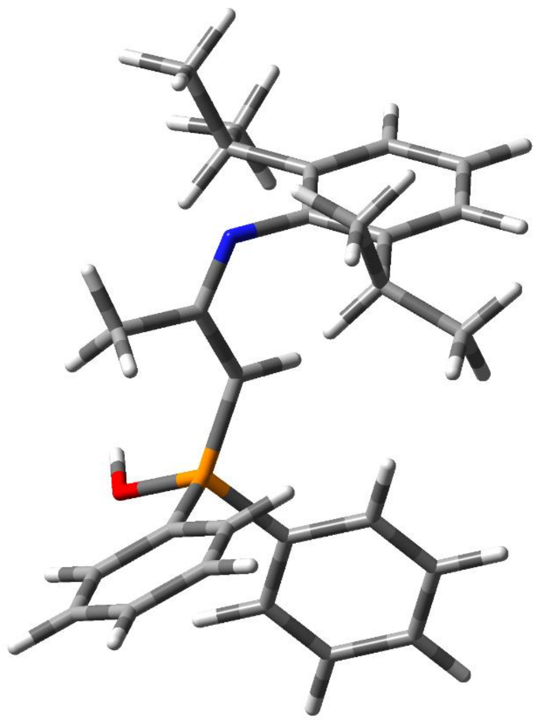
(E)-enamine 2, Energy: --1518.98068 Hartrees



P	1.717000	0.460300	-1.143900
O	1.939900	0.809300	-2.576800
N	-1.881600	-1.323000	-0.747900
C	0.064400	0.057100	-0.652800
H	-0.416100	0.723600	0.053000
C	-0.615800	-1.006000	-1.145800
C	-0.038200	-1.979900	-2.132900
H	0.262200	-2.891900	-1.612100
H	0.825200	-1.569100	-2.649500
H	-0.787800	-2.248600	-2.879000
C	-2.655100	-0.529000	0.148100
C	-2.414700	-0.614800	1.523600
C	-3.196000	0.162700	2.375600
H	-3.032000	0.109000	3.445200
C	-4.182900	0.995000	1.876800
H	-4.783300	1.589100	2.554000
C	-4.398900	1.076200	0.508900
H	-5.164300	1.740900	0.131000
C	-3.634200	0.327600	-0.378900
C	-1.357800	-1.533300	2.102600
H	-0.819000	-1.990400	1.272600
C	-2.004900	-2.656800	2.916200
H	-2.701500	-3.232800	2.305700
H	-2.556300	-2.250400	3.766500
H	-1.241800	-3.335000	3.302100
C	-0.349500	-0.759200	2.953500
H	0.092300	0.062300	2.387000
H	0.453000	-1.420500	3.287000
H	-0.823100	-0.339300	3.842900
C	-3.864200	0.397600	-1.875100

H	-2.901000	0.218300	-2.358700
C	-4.834700	-0.704800	-2.314500
H	-4.490200	-1.698600	-2.023200
H	-4.963700	-0.693400	-3.397900
H	-5.811100	-0.546300	-1.851400
C	-4.370200	1.757700	-2.348200
H	-5.392500	1.943900	-2.014400
H	-4.373000	1.788800	-3.438300
H	-3.736800	2.567100	-1.983700
C	2.783100	-0.924600	-0.646900
C	2.467500	-1.772000	0.413400
H	1.539100	-1.630600	0.955900
C	3.328300	-2.802700	0.765700
H	3.077000	-3.458700	1.589200
C	4.506400	-2.996300	0.055300
H	5.175200	-3.803000	0.326800
C	4.822600	-2.159900	-1.007900
H	5.736600	-2.314800	-1.566500
C	3.964000	-1.127000	-1.358100
H	4.195200	-0.477400	-2.193100
C	2.170300	1.832600	-0.048700
C	2.388600	1.657600	1.316600
H	2.327700	0.668400	1.755700
C	2.704400	2.745100	2.117400
H	2.876200	2.603700	3.176500
C	2.807800	4.013300	1.557200
H	3.056600	4.861100	2.182400
C	2.599800	4.191200	0.196500
H	2.686800	5.177000	-0.241600
C	2.282800	3.102500	-0.606400
H	2.126700	3.226600	-1.670600
H	-2.375300	-1.995800	-1.313200

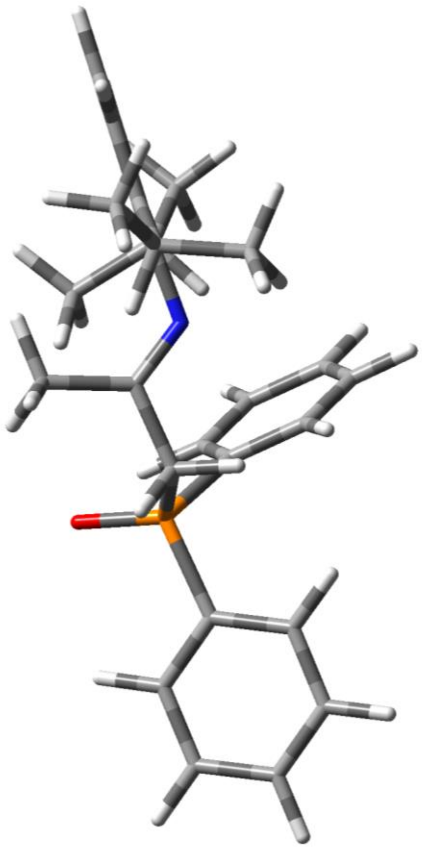
(Z)-ylide, Energy: -1518.946878 Hartrees



P	1.625100	0.322300	-0.959800
O	2.101500	0.541500	-2.488600
N	-1.920300	-1.436500	-0.583400
C	0.006200	0.010700	-0.658300
H	-0.543200	0.839900	-0.232100
C	-0.693300	-1.217800	-0.914200
C	0.031400	-2.357400	-1.588400
H	0.700700	-2.847200	-0.877000
H	0.640700	-2.014200	-2.426800
H	-0.693200	-3.087400	-1.941200
C	-2.639200	-0.452500	0.117700
C	-2.535800	-0.375600	1.520000
C	-3.317800	0.555000	2.197400
H	-3.251000	0.627700	3.275400
C	-4.180000	1.400700	1.514900
H	-4.779300	2.120300	2.058200
C	-4.263200	1.326100	0.132800
H	-4.928000	1.998900	-0.393500
C	-3.499800	0.409900	-0.584800
C	-1.616200	-1.330600	2.257100
H	-0.716300	-1.446400	1.648000
C	-2.277000	-2.707800	2.379300
H	-2.549200	-3.093500	1.397200
H	-3.181800	-2.634600	2.987600
H	-1.600900	-3.419000	2.858700
C	-1.181600	-0.830300	3.631500
H	-0.765900	0.178000	3.581900
H	-0.420200	-1.495000	4.042900

H	-2.015600	-0.817400	4.336300
C	-3.597300	0.293700	-2.093200
H	-2.590000	0.086700	-2.463800
C	-4.484900	-0.897700	-2.468600
H	-4.112600	-1.813800	-2.011000
H	-4.513100	-1.032900	-3.551900
H	-5.505900	-0.724300	-2.120000
C	-4.096600	1.561900	-2.778400
H	-5.146400	1.754900	-2.547400
H	-4.018000	1.452400	-3.861100
H	-3.516900	2.436800	-2.479100
C	2.776800	-0.981000	-0.485100
C	2.661100	-1.500900	0.803700
H	1.885700	-1.130600	1.465600
C	3.522200	-2.498900	1.230200
H	3.427400	-2.902500	2.229600
C	4.500400	-2.984500	0.369800
H	5.169400	-3.768300	0.700500
C	4.618400	-2.467800	-0.912700
H	5.378300	-2.847700	-1.583000
C	3.760600	-1.463600	-1.343000
H	3.848800	-1.064100	-2.343800
C	2.099200	1.825900	-0.090300
C	1.511100	2.157000	1.130200
H	0.735100	1.526300	1.545900
C	1.919300	3.295300	1.809600
H	1.456500	3.551800	2.753300
C	2.917500	4.102400	1.279500
H	3.234200	4.990000	1.811600
C	3.510200	3.771900	0.067800
H	4.287900	4.400100	-0.346100
C	3.104700	2.635600	-0.616800
H	3.564600	2.378600	-1.562500
H	1.433500	1.007200	-3.009100

(E)-imine 2, Energy: -1518.983201 Hartrees



P	2.016800	-0.206900	-0.716000
O	1.943300	0.074000	-2.174800
N	-1.277200	-0.591700	0.319500
C	0.865100	-1.508500	-0.168300
H	0.954700	-1.640500	0.909800
C	-0.557000	-1.175300	-0.544200
C	-0.967400	-1.569100	-1.934300
H	-0.964600	-2.659200	-2.015800
H	-0.227900	-1.189700	-2.642900
H	-1.955800	-1.193100	-2.188800
C	-2.628500	-0.263200	0.061300
C	-2.957700	1.069900	-0.224000
C	-4.299800	1.392100	-0.408000
H	-4.576200	2.413600	-0.634300
C	-5.289500	0.429100	-0.304300
H	-6.327700	0.697900	-0.451500
C	-4.946900	-0.881000	-0.004300
H	-5.728400	-1.624200	0.079000
C	-3.620400	-1.253000	0.187300
C	-1.868400	2.114700	-0.371600
H	-1.065600	1.849600	0.321800
C	-2.334200	3.527700	-0.028100
H	-2.826900	3.563500	0.944900
H	-3.026500	3.917500	-0.777200

H	-1.471900	4.195600	-0.000800
C	-1.291700	2.086900	-1.791300
H	-0.779800	1.147400	-2.006300
H	-0.568000	2.893300	-1.923200
H	-2.091500	2.220200	-2.524100
C	-3.242500	-2.670200	0.578800
H	-2.348300	-2.947000	0.015600
C	-2.888000	-2.724000	2.069200
H	-2.091500	-2.020000	2.310300
H	-2.563900	-3.727800	2.350600
H	-3.765400	-2.468200	2.667500
C	-4.315300	-3.705600	0.256700
H	-5.199300	-3.571900	0.883200
H	-3.927800	-4.706700	0.449600
H	-4.625900	-3.653900	-0.787900
C	3.647800	-0.802700	-0.215300
C	3.958600	-1.102600	1.110500
H	3.220500	-0.970600	1.892900
C	5.222900	-1.565900	1.439200
H	5.461900	-1.797000	2.468900
C	6.182700	-1.729100	0.446100
H	7.170100	-2.089000	0.705000
C	5.877600	-1.429500	-0.874000
H	6.625400	-1.554500	-1.646100
C	4.610700	-0.967000	-1.206600
H	4.357500	-0.727900	-2.231700
C	1.647400	1.247600	0.287300
C	1.269000	1.182100	1.627700
H	1.156000	0.228400	2.126300
C	0.988900	2.345300	2.328600
H	0.684500	2.288400	3.365300
C	1.080700	3.578700	1.694800
H	0.850600	4.484500	2.241000
C	1.457200	3.649300	0.360500
H	1.522300	4.608700	-0.136400
C	1.741000	2.486500	-0.342100
H	2.016900	2.527300	-1.388500
H	1.199800	-2.421700	-0.665100

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