

Preparation of $\text{Ta}_2\text{Cp}'_2\text{Cl}_4(\text{H})(\text{O})(\text{CHPMe}_3)$

PMe_3 (0.2 ml, 2.6 mmol) was added to a solution of $\text{Ta}_2\text{Cp}'_2\text{Cl}_4\text{H}(\text{CHO})$ (0.83 g, 1 mmol) in 50 ml of benzene. The color changed from orange to yellow in 0.5 h. Solvent was removed under vacuum, leaving crude, yellow $\text{Ta}_2\text{Cp}'_2\text{Cl}_4\text{H}(\text{O})(\text{CHPMe}_3)$. Recrystallization from toluene/pentane gave 0.76 g (84%) yellow crystals in two crops.

Anal. Calcd. for $\text{Ta}_2\text{C}_{26}\text{H}_{45}\text{Cl}_4\text{OP}$: C, 34.38; H, 4.99. Found: C, 34.47; H, 5.10. ^1H NMR (CDCl_3): ppm 9.84 (dd, 1, $J_{\text{HH}} = 5$ Hz, $J_{\text{HP}} = 2$ Hz, TaH), 2.47 (m, 1, $\text{C}_5\text{CH}_2\text{CH}_3$), 2.44 (m, 1, $\text{C}_5\text{CH}_2\text{CH}_3$), 2.21 (d, 2, $\text{C}_5\text{CH}_2\text{CH}_3$), 2.19-2.13 (6 singlets, 24 total, C_5CH_3), 1.69 (d, 9, $J_{\text{HP}} = 12$ Hz, PMe_3), 1.44 (dd, 1, $J_{\text{PH}} = 13$ Hz, $J_{\text{HH}} = 5$ Hz, CHPMe_3), 1.05 (t, 3, $\text{C}_5\text{CH}_2\text{CH}_3$), 0.99 (t, 3, $\text{C}_5\text{CH}_2\text{CH}_3$). $^{31}\text{P}\{^1\text{H}\}$ NMR (CHCl_3): 22.5 ppm (s).

$\text{Ta}_2\text{Cp}'_2\text{Cl}_4\text{H}(\text{O})(^{13}\text{CHPMe}_3)$ was prepared similarly from $\text{Ta}_2\text{Cp}'_2\text{Cl}_4\text{H}(^{13}\text{CHO})$ and PMe_3 in benzene. ^{13}C NMR (CDCl_3 , gated, proton decoupled): ppm 94.13 (ddd, $J_{\text{CP}} = 44$ Hz, $J_{\text{CH}} = 122$ Hz, $J_{\text{CH}} = 10$ Hz, CHPMe_3).

Preparation of $[\text{Ta}(\eta^5\text{-C}_5\text{Me}_4\text{R})\text{Cl}_2\text{H}]_2(\text{py})$

Pyridine (1.48 mmol) was added to an incompletely dissolved sample of $[\text{TaCp}''\text{Cl}_2\text{H}]_2$ (0.59 g) in 50 ml of toluene. The starting material dissolved to give a deep green solution which was filtered and concentrated to \sim 35 ml. Pentane (\sim 15 ml) was added and the reaction was cooled to -30° . The product crystallized as dark green, almost black, crystals (0.6 g, 92%). It is less soluble than the starting material (in the absence of excess pyridine) in benzene or toluene. Pyridine of crystallization can be observed in the NMR spectra of most samples.