

Tantalum-Catalyzed Hydroaminoalkylation: a DFT investigation on a non-innocent ligand

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Hydroaminoalkylation (HAA) is a very efficient catalytic method for alkene and alkyne functionalization with amines [1]. Tantalum complexes bearing 1,3-N,O ligands have proved to be highly effective catalysts in this transformation (Figure 1) [2]. Although some steps of the reaction mechanism have been previously studied [3], the role of the ligand has not been fully clarified.

In this study, a combined experimental and computational investigation reveals how the ligand actively participates in the reaction as a proton shuttle during the whole mechanism, lowering the energy of transition states and enabling the high activity of the catalyst. The new proposed mechanism is also able to rationalize why 1,4- and 1,5-N,O-chelating ligands suppress the catalyst reactivity, enhancing the bond strength to the metal.

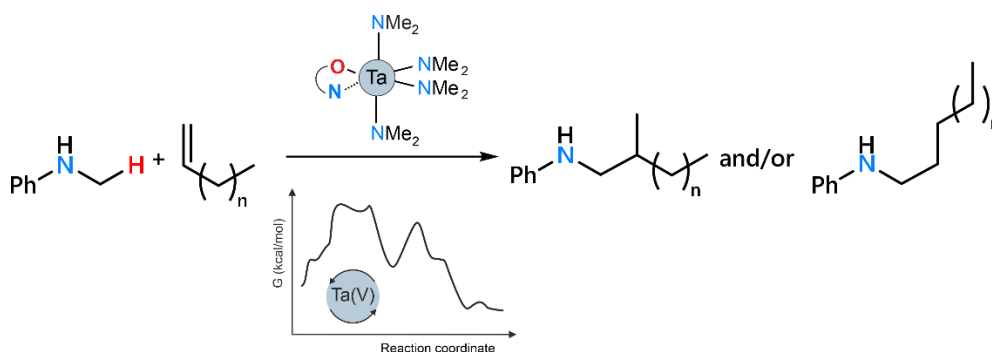


Figure 1. Hydroaminoalkylation catalyzed by tantalum complexes bearing 1,n-N,O chelating ligands.

References

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