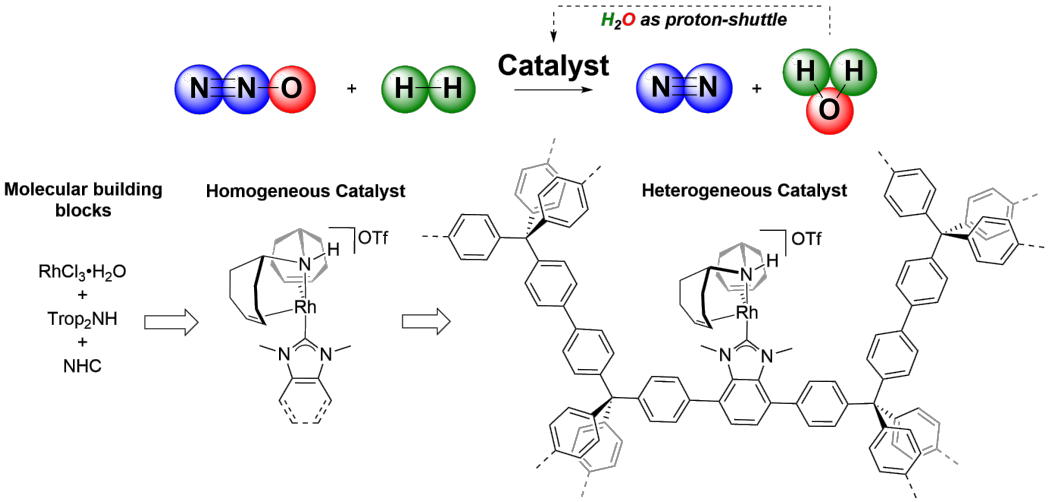
Metal-ligand Cooperative Rhodium Complexes as Highly Active N_2O Hydrogenation Catalysts

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Nitrous oxide (N_2O), more commonly known as laughing gas, is no laughing matter in view of its significance as an atmospheric pollutant. In recent times, N_2O has been identified as the most ozone depleting agent of the 21st century (comparable to that of CFC's) and a potent greenhouse gas.[1] The 'simple on-paper' reaction: $N_2O + H_2 \rightarrow N_2 + H_2O$ can be a promising mitigation strategy, but in reality this reaction is difficult to perform. Activation of N_2O is challenging, in addition, the catalyst must remain robust and selective towards the oxidizing (N_2O) and reducing (H_2) conditions. N_2O have been, and generally still is, broadly overlooked compared to its isoelectronic and isostructural more well known molecule, CO_2. In the past decades we have seen strong research forces yielding several CO_2 hydrogenation catalysts with turnover numbers (TON's) up to millions.[2] Meanwhile the homogeneous N_2O hydrogenation catalysts in the scientific literature today are very few in numbers and only reaches TON's of hundreds.[3]



We can now report that we have found a catalyst that shows unprecedented activity (~ 200'000 TON's) in the hydrogenation of N_2O at mild conditions (2-5 bars, 65 °C). Rhodium as the metal and NHC's as ancillary ligands combined with a tridentate bis-olefine amine ligand[4] was found to be the best among the tested candidates. The credit cannot be given to the catalyst design alone as evidence suggests a more holistic process. Our combined experimental and theoretical study of the reaction mechanism concludes that the process is autocatalytic. In a bottom-up approach the catalyst could be heterogenized, and the activity and stability of the well-defined single-site heterogeneous catalyst was evaluated and compared with the molecular version.

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[4] Thomas L. Gianetti, Hansjörg Grützmacher, et. al., *Angewandte Chemie*, **2016**, 55, 1854-1858. ; and literature cited therein.