Computational Design of Active Site Structures with Improved Transition-State Scaling for Ammonia Synthesis - DTU Orbit (07/10/2019)

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The Haber-Bosch process for the reduction of atmospheric nitrogen to ammonia is one of the most optimized heterogeneous catalytic reactions, but there are aspects of the industrial process that remain less than ideal. It has been shown that the activity of metal catalysts is limited by a Brønsted-Evans-Polanyi (BEP) scaling relationship between the reaction and transition-state energies for N$_2$ dissociation, leading to a negligible production rate at ambient conditions and a modest rate under harsh conditions. In this study, we use density functional theory (DFT) calculations in conjunction with mean-field microkinetic modeling to study the rate of NH$_3$ synthesis on model active sites that require the singly coordinated dissociative adsorption of N atoms onto transition metal atoms. Our results demonstrate that this "on-top" binding of nitrogen exhibits significantly improved scaling behavior, which can be rationalized in terms of transition-state geometries and leads to considerably higher predicted activity. While synthesis of these model systems is likely challenging, the stabilization of such an active site could enable thermochemical ammonia synthesis under more benign conditions.

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