

Towards Computational Design of Single Transition Metal Atom Catalysts for Nitrogen Fixation

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Abstract: Ammonia (NH₃) plays an essential role in agriculture to meet global population explosion. Although the atmosphere consists of more than 78% of Nitrogen (N₂), industrial NH₃ synthesis from atmospheric N₂ gas requires harsh conditions of temperature (700~850 K) and pressure (50~200 atm), which annually consuming 2% of the world's power. Recently, an electrocatalytic NH₃ fixation process under ambient condition was introduced. With this, six protons and electrons are gradually added ($6\text{H}^+ + 6\text{e}^- + \text{N}_2 = 2\text{NH}_3$), instead of breaking the triple bonds of N₂ in the traditional method, thus saving dramatic amount of energy. Along this theme, the synthesis of NH₃ under ambient condition has become one of the most attractive and challenging chemical process. In particular, the search for appropriate catalyst is obviously an important task. It is known that single metal atom supported on a two-dimensional monolayer has highly promising catalytic properties such as hydrogen evolution reaction, oxygen evolution reaction and CO reduction reaction. To complement this, computational tools such as density functional theory (DFT) have been successfully used to speed up the design of new functional materials for a targeted application. In this work, we use DFT calculation to search for new nano-scale functional materials for N₂ fixation with thermal stability and high catalytic activity, and to explain the heterogeneous catalytic mechanism on the surface of the catalyst at atomic scale. The feasibility of single transition metal atoms (Sc to Zn, Mo, W, Ru, Pd, Pt, Ag and Au) supported on experimentally available phosphorene monolayer was systematically screened for N₂ fixation. In addition, the catalytic mechanism and nitrogen reduction reaction steps were also studied. The results show that single transition metal supported on phosphorene exhibits high catalytic activity for N₂ fixation at room temperature with fairly low overpotential. It will be shown in the presentation that the N-N triple bond will be weakened by back-donation from the transition metal to N₂ molecule.