A theoretical evaluation of possible transition metal electro-catalysts for N₂ reduction - DTU Orbit (07/12/2018)

A theoretical evaluation of possible transition metal electro-catalysts for N₂ reduction

Theoretical studies of the possibility of forming ammonia electrochemically at ambient temperature and pressure are presented. Density functional theory calculations were used in combination with the computational standard hydrogen electrode to calculate the free energy profile for the reduction of N₂ admolecules and N adatoms on several close-packed and stepped transition metal surfaces in contact with an acidic electrolyte. Trends in the catalytic activity were calculated for a range of transition metal surfaces and applied potentials under the assumption that the activation energy barrier scales with the free energy difference in each elementary step. The most active surfaces, on top of the volcano diagrams, are Mo, Fe, Rh, and Ru, but hydrogen gas formation will be a competing reaction reducing the faradaic efficiency for ammonia production. Since the early transition metal surfaces such as Sc, Y, Ti, and Zr bind N-adatoms more strongly than H-adatoms, a significant production of ammonia compared with hydrogen gas can be expected on those metal electrodes when a bias of 1 V to 1.5 V vs. SHE is applied. Defect-free surfaces of the early transition metals are catalytically more active than their stepped counterparts.

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