

Electroreduction of N₂ to ammonia at ambient conditions on mononitrides of Zr, Nb, Cr, and V – A DFT guide for experiments - DTU Orbit (08/11/2017)

Electroreduction of N₂ to ammonia at ambient conditions on mononitrides of Zr, Nb, Cr, and V – A DFT guide for experiments

A rapid and facile reduction of nitrogen to achieve a sustainable and energy efficient production of ammonia is critical to its use as a hydrogen storage medium, chemical feedstock and especially for manufacturing inorganic fertilizers. For a decentralization of catalytic ammonia production, small-scale N₂ reduction devices are required that are equipped with the most stable, selective and active catalysts that operate at low temperature and ambient pressure. Here, we report the development of new and cost-efficient catalysts, transition metal nitrides, which enable electrochemical reduction of molecular nitrogen to ammonia in aqueous media at ambient conditions with only a low applied bias. The most promising catalysts are VN, ZrN, NbN and CrN, which are identified among a range of transition metal nitride surfaces through a comprehensive density functional theory based analysis. All four nitrides are found to be more active towards nitrogen reduction than towards the competing hydrogen evolution reaction, in contrast to pure metal catalysts, which largely evolve hydrogen. Furthermore, their stability against poisoning and possible decomposition under operating conditions is also studied. Particular single-crystal surfaces are needed for ZrN, NbN and CrN because polycrystalline surfaces may result in decomposition of the whole catalyst. Polycrystalline surfaces of VN may, however, be used since the rocksalt (100) facet is predicted to produce ammonia via a Mars-van Krevelen mechanism with only a -0.5 V overpotential, thereby avoiding decomposition. We suggest that this is a promising step towards the development of a method for synthesizing ammonia cheaply, to prepare high-value-added nitrogenous compounds directly from air, water and electricity at ambient conditions. An additional benefit to the present analysis is that the method used in this work may be applicable to other aqueous phase catalytic reactions, where a Mars-van Krevelen mechanism is operative and product selectivity and activity are key catalytic criteria.

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