Two-state Reaction Mechanism of the Synthesis of Ammonia in the N2/H2/Ru Systems: A Theoretical Study

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Abstract

Ru-based catalysts show high activity and stability to produce ammonia. Herein, the two-state reaction mechanism of Ru catalyzes N2 and H2 to synthesize NH3 are theoretical studied with the density functional theory (DFT) UB3LYP methods. The spin-orbital coupling constant (Hsoc) and intersystem crossing probability (Pisc) at minimum energy crossing point (MECP) were calculated, respectively. Its are: Hsoc, MECP1 = 508.34 cm⁻¹, P2, MECP1 ISC = 0.85, MECP2: Hsoc, MECP2 = 269.21 cm⁻¹, P2, MECP2 ISC = 0.27. Used energy span model to determined TOF-determining transition state (TDTS) as 3TS2-3 and TOF-determining intermediate (TDI) as 3IM9 of reaction. In addition, the charge decomposition analysis (CDA), spin population analysis and frontier molecular orbital (FMO) theory were used to analyzed reaction mechanism.

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