

Developing and Unraveling the Active Sites of Ruthenium-based Catalysts for Mild Ammonia Synthesis

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ABSTRACT

The development of efficient and stable Ru catalysts with favorable textural properties is crucial for synthesis of decarbonized NH₃, a potential hydrogen carrier. Herein, a series of γ -Al₂O₃-supported Cs-promoted Ru catalysts were prepared, among which a 1.5Cs-Ru/ γ -Al₂O₃ catalyst with a Cs/Ru molar ratio of 1.5 and a Ru size of ~2 nm exhibited high NH₃ synthesis rates (7-30 mmolNH₃ g⁻¹ h⁻¹) at 410 °C and 1 MPa. In contrast to the previous speculation regarding tuning the electron environment of Ru using CsOH, this study unraveled that tuning Cs/Ru molar ratio not only varied surface acidity/basicity but also generated new active sites at the interfaces of Ru particles with CsOH-Cs⁰ species for ammonia synthesis. Among the active sites of this 1.5Cs-Ru/ γ -Al₂O₃ catalyst, Cs⁰ (minor), which transferred electrons to Ru, was present at the boundaries of Ru particles and CsOH (major), that attracted the dissociative hydrogen atoms from the metallic Ru surfaces, via spillover effect, in addition to that changed surface acidity/basicity, facilitating the adsorption/adsorption of nitrogen and ammonia, and thus promoting NH₃ synthesis. The results provide insights into the active sites and corresponding functions for NH₃ synthesis over alkali-promoted metal catalysts, which are readily applicable to a wide range of industrial hydrotreating reactions.

