Revealing Key Intermediates on Ru catalysts in Ammonia Synthesis by in situ Infrared Spectroscopy

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NH_3 is one of the major candidates for a carbon-neutral energy carrier to realize a sustainable society. Present industrial NH_3 synthesis via the Haber-Bosch process using Fe-based catalysts requires harsh conditions of high temperature and pressure. On the other hand, Ru-based catalysts exhibit higher activity under milder conditions than Fe-based catalysts.[1] This enhanced activity is attributed to the efficient generation of reaction intermediates. Consequently, understanding the activated adsorption states and reaction intermediates on Ru-based catalysts is crucial for elucidating the efficient NH_3 synthesis mechanism, and contributes to the design of the noble catalysts that exhibit higher activity under milder conditions. In this study, we aim to elucidate the adsorption states of the reactants, N_2 and H_2, and the formation of surface intermediates on Ru catalysts under reaction conditions using in situ FT-IR combined with modulation excitation spectroscopy (IR-MES) measurements.[2]

We prepared Ru nanoparticles supported on MgO (Ru/MgO) with a Ru-loading of 4-5 wt% as the sample for in situ FT-IR measurements. The in situ FT-IR spectra at 250 °C were obtained under the reaction gas (0.1 MPa, a N_2-H_2 mixture gas) after N_2 pre-adsorption. This in situ spectra exhibited the time-domain absorbance peaks attributed to the vibrations of pre-adsorbed N_2, H atoms on Ru, NH_3, and a horizontally adsorbed N_2H intermediate. This result suggests that the horizontal adsorption of N_2H is the only possible intermediate for the NH_3 reaction under this condition, i.e., we consider that the horizontal adsorption state of N_2H plays an important role in the activation of N_2 under these conditions. The in situ observation of these intermediates provides insight into key steps for NH_3 synthesis on Ru catalysts under milder conditions and contributes to the design of the noble metal catalysts for NH_3 synthesis.

References

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