

International Institute for Carbon-Neutral Energy Research (WPI-I<sup>2</sup>CNER) Kyushu University

## Title Design of core-shell structured metal catalysts for synthesis of carbon-free ammonia fuel

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### Abstract

Date

Ammonia has attracted great attenuations as a H<sub>2</sub> (energy) carrier for utilizing renewable energy and a carbon-free fuel because CO<sub>2</sub> is not emitted when ammonia is decomposed to produce H<sub>2</sub> or combusted in power station. Ammonia is commonly produced by the Haber-Bosch process. In this process, ammonia is produced at high temperatures (>450 °C) and pressures (>20 MPa) over an iron-based catalyst and the use of fossil fuels emits large amount of CO<sub>2</sub>. A catalyst that yields ammonia with high efficiency under mild reaction conditions (<400 °C, <10 MPa) would reduce energy consumption in the green ammonia fuel synthesis process.

Ru-based catalysts shows higher activity than iron-based catalyst under mild reaction conditions. However, active Ru catalyst loaded on oxide support, which is easily prepared and stable in air and thus suitable for practical application, is yet to be developed. We reported that composite-rare-earth oxides such as  $La_{0.5}Ce_{0.5}O_{0.75}$  and  $La_{0.5}Pr_{0.5}O_{1.75}$  supported Ru catalysts showed high ammonia synthesis rate under mild conditions (300 - 400 °C, 1.0 MPa), when the catalysts were pre-reduced at unusually high temperature, 650 °C. Furthermore, we found that Ru/Ba<sub>0.1</sub>La<sub>0.45</sub>Ce<sub>0.45</sub>O<sub>1.68</sub> reduced at 700 °C showed the comparable to or better activity than other reported oxide supported Ru catalysts.

Such design concept of catalyst was applied to development of Co catalysts. We discovered that Co/BaO/MgO reduced at 700 °C exhibited higher ammonia synthesis rate than bench-mark Ru catalysts at 350 °C and 1.0 MPa. At the same temperature the ammonia synthesis rate of Co/BaO/MgO was increased drastically with increase in pressure to 3.0 MPa and was higher than that of active Ru-based catalyst such as Ru/  $La_{0.5}Ce_{0.5}O_{0.75}$ . This is because Co/BaO/MgO is free from hydrogen poisoning, which is typical drawback for Ru catalysts. It was uncovered that the Co nanoparticles are encapsulated by nano-fractions of BaO and all the surface-available Co sites contact with the low-crystalline nano-fractions, and electrons are effectively donated from the nano-fractions *via* Co to antibonding  $\pi$ -orbital of N<sub>2</sub>, which results in weakening of N=N bond, rate determining step for ammonia synthesis.

#### About the Speaker

Katsutoshi Nagaoka received his Ph. D. at Tokyo Institute of Technology in 2001. After Post Doctor in the university and Humboldt fellow in Technical University in München, he moved to Oita University as Lecturer in 2004 and he was promoted to Associate Professor the next year. In 2019 he was appointed to full professor in Nagoya University. His research interests involve heterogeneous catalysis for renewable energy utilization and elements recycling to solve global energy and environmental problems. He was awarded The Japan Petroleum Institute Prize for Encouragement (2009) and Catalysis Society of Japan Prize for Encouragement (2010).

Registration https://zoom.us/webinar/register/WN\_TKbXaPMbTsWFauqOSKEFhw

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