

## Development of Nano-nickel Catalyst by Using Supercritical CO<sub>2</sub> for Methane Cracking

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### Extended Abstract

Hydrogen is highly desirable for use in proton-exchange membrane fuel cells. Production of hydrogen can be achieved via the direct cracking of methane over supported nickel catalysts. The products of methane cracking reaction are CO-free hydrogen and solid carbon, therefore it is not required to purify the obtained hydrogen before use. However, the major problem of this process is the catalyst deactivation due to carbon formation over the catalyst surface [1]. Thus, it is necessary to prepare supported nickel nanoparticles with the desirable particle size and distribution in order to prolong the catalyst lifetime. Using supercritical fluid to prepare catalysts has been receiving attention as an alternative method. The unique properties of supercritical fluids such as gas-like diffusivity and viscosity, low surface tension, high compressibility combined with liquid-like density make these fluids suitable for impregnation of active metals into a porous support [2-3].

In this study, various preparation methods of nano-Ni/SBA-15 catalyst were developed by using supercritical carbon dioxide with ethanol as a co-solvent (SCCO<sub>2</sub>-EtOH) and Gas Anti Solvent (GAS) techniques. The prepared catalysts were then used for methane cracking reaction. The effects of nickel loading, pressure, temperature and amount of co-solvent on the catalyst size and size distribution, hydrogen uptake, catalyst activity, amount and type of carbon formed were investigated. The obtained results were compared to the Ni/SBA-15 catalyst prepared by incipient wetness impregnation. It was found that the catalysts prepared by SCCO<sub>2</sub>-EtOH method had better size distribution, smaller particle size and higher hydrogen uptake compared to the incipient wetness impregnation. For the SCCO<sub>2</sub>-7.33 mol% EtOH method, it was found that an increase in pressure resulted in a better dispersion of nickel metals inside the porous SBA-15 support. In addition, an increase in temperature caused a reduction in catalyst size. In the catalytic activity studies, it was found that at the beginning of the reaction the catalyst prepared by the incipient wetness impregnation method had a higher activity but deactivated faster compared to those prepared by the SCCO<sub>2</sub>-EtOH method. The catalysts prepared by the GAS method had the lowest catalytic activity and shortest lifetime, whereas the catalyst prepared by the SCCO<sub>2</sub>-7.33 mol% EtOH method at 120 bar and 45 °C had the highest catalytic activity. In addition, it was found that increasing the nickel metal loading could prolong the lifetime of the catalyst. In the studies of amount and type of carbon formed, three types of carbon, namely, amorphous carbon, less-stable Single-Walled Carbon Nanotube (SWCNT) and carbon nano-fiber were observed at the end of the reaction for all catalyst preparation methods. As the amount of nickel was increased, the amount of carbon formed and the catalytic performance were also increased. For tests in which a reduced amount of ethanol was used as a co-solvent (2.5 mol%) to prepare catalysts, it was found that a reduction in the amount of co-solvent had no significant effect on the particle size and size distribution.

### References

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