Pornprapa Saithongsuk 2014: Development of Nano-nickel Catalyst by Using Supercritical Carbon Dioxide with Ethanol for Methane Cracking Reaction. Master of Engineering (Chemical Engineering), Major Field: Chemical Engineering, Department of Chemical Engineering. Thesis Advisor: Associate Professor Manop Charoenchaitrakool, Ph.D. 123 pages.

Various preparation methods of nano-Ni/SBA-15 catalyst were developed by using supercritical carbon dioxide with ethanol as a co-solvent ($SCCO_2$-EtOH) and Gas Anti Solvent (GAS) techniques. The prepared catalysts were then used for methane cracking reaction. In this study, 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 $SCCO_2$-EtOH method had better size distribution, smaller particle size and higher hydrogen uptake compared to the incipient wetness impregnation. For the $SCCO_2$-7.33 mol% EtOH method, it was found that an increase in pressure resulted in a better dispersion of catalysts inside the porous 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 shorter lifetime compared to those prepared by the $SCCO_2$-EtOH method. The catalysts prepared by the GAS method had the lowest catalytic activity and shortest lifetime, whereas the catalyst prepared by the $SCCO_2$-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 SWCNTs 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.

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