Bi-reforming of Methane with Steam and CO_2 over Ni/La-SBA-15 Catalyst for Synthesis Gas Production

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Bi-reforming of methane with steam and CO_2 has been emerged as an alluring reforming technique for syngas production with a desirable H_2/CO ratio close to 2, which is highly compatible for the industrial generation of methanol and Fischer-Tropsch chemicals. This study focuses on preparation, physicochemical attributes and the catalytic performance of 10%Ni/La-SBA-15 catalyst for the combined steam and dry reforming of methane. The La_2O_3-incorporated mesoporous silica support (La-SBA-15) previously prepared by one-pot hydrothermal method was doped with $Ni(NO_3)_2$ precursor solution using an incipient wetness impregnation approach to synthesize a 10%Ni/La-SBA-15 catalyst. Various characterization techniques including XRD, FTIR, BET, HR-TEM, H_2-TPR and TPO measurements were employed to investigate the physicochemical properties of both fresh and spent catalysts. Bi-reforming of methane was carried out in a tubular fixed-bed reactor under atmospheric pressure at 923 K – 1023 K and CH_4:CO_2:H_2O = 2:1:1 with gas hourly space velocity (GHSV) of 36 L g_{cat}^{-1} h^{-1}. Although La-SBA-15 support possessed high BET surface area of 737.3 m^2 g^{-1}, an unavoidable decline in surface area with Ni metal addition to 535.4 m^2 g^{-1} for 10%Ni/La-SBA-15 catalyst was evident indicating the diffusion of NiO nanoparticles into the mesopores of La-SBA-15 support. Indeed, H_2-TPR result shows that NiO particles present on support surface were reduced to metallic Ni^0 phase at 675 K, while NiO species inhabited inside the mesoporous channels of support were completely reduced at higher reduction temperature of above 750 K due to strong metal-support interaction. Additionally, the presence of NiO phase with small average crystallite size of 17.4 nm was confirmed by X-ray diffraction measurement. The 10%Ni/La-SBA-15 catalyst exhibited stable activity and selectivity during the span of 12 h on-stream regardless of reaction temperature of 923-1023 K. Methane conversion increased with growing reaction temperature from 923 to 1023 K and reached to the highest value of about 55%. Interestingly, H_2/CO ratio also enhanced with rising temperature and it was always greater than 1. In addition, H_2 selectivity and yield were superior to those of CO. These observations would be due to the concomitant presence of CH_4 steam reforming reaction favored for H_2 formation.