ETHANOL DRY REFORMING FOR H₂-RICH SYNGAS PRODUCTION OVER Ce-Ni/Al₂O₃ CATALYST

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ABSTRACT

The transformation of greenhouse gas, CO₂ to fuels using heterogeneously catalytic processes has received significant attention due to the depleting petroleum resources, associated environmental problems and stringent environmental regulations. CO₂ reforming of CH₄ is considered as a suitable method for producing syngas to substitute petroleum-based energy. However, CH₄ is an unrenewable resource and will probably be depleted next century. Thus, there is requirement of an alternatively sustainable and green approach for H₂ synthesis. The production of H₂ from ethanol dry reforming seems to be a promising method since it utilizes both bio-derived ethanol regarded as renewable resource and undesirable CO₂ as feedstocks. However, the knowledge about CO₂ reforming of ethanol is still limited. Hence, the aim of this research was to examine the physicochemical properties of 3%Ce-10%Ni/Al₂O₃ catalyst and study the influence of both reaction temperature and reactant partial pressure on activity and selectivity of CO₂ reforming of ethanol. Co-impregnation method between Ce(NO₃)₃ and Ni(NO₃)₂ precursor solutions and γ-Al₂O₃ support thermally pretreated at 973 K was used for synthesizing 3%Ce-10%Ni/Al₂O₃ catalyst. Ethanol dry reforming runs were carried out in a quartz fixed-bed reactor placed vertically in a split tubular furnace at reaction temperature range of 873 - 973 K under atmospheric pressure. Approximately 0.1 gcat of catalyst was sandwiched by quartz wool in the middle of the reactor. Gas hourly space velocity, GHSV= 42 L gcat⁻¹ h⁻¹ and mean particle size of 100-140 µm were used for all runs to ensure that the negligible internal and external transport resistances was present. The ratios of CO₂ to C₂H₅OH were varied from 1:1 to 2.5:1 and vice versa. CO₂ and C₂H₅OH mixture was diluted in N₂ flow acting as a tie component for material balance purposes and ensuring the total flowrate of 70 ml min⁻¹. 3%Ce-10%Ni/Al₂O₃ catalyst possesses high surface area of 138.56 m² gcat⁻¹ with pore volume and pore diameter of 0.23 cm³ gcat⁻¹ and 59 nm, respectively. The BET area of catalyst was close to that of calcined γ-Al₂O₃, NiO, CeO₂ and NiAl₂O₄ phases were formed on catalyst surface during calcination. Both CO₂ and C₂H₅OH conversions initially decreased with time-on-stream and appeared to be stable at ca. 4-5 h. A reduction in H₂/CO ratio from 20 to 5 was observed with increasing reaction temperature from 873 K to 973 K probably due to the enhancement of reverse water-gas shift reaction. The consumption rate of C₂H₅OH and CO₂ was improved with increasing partial pressure of CO₂, P_CO₂. Interestingly, C₂H₅OH and CO₂ conversions also enhanced with growing P_C₂H₅OH and approached the maximum at around 30-35 kPa.

Keywords: Ni-based catalyst; Ethanol dry reforming; Hydrogen; Syngas; Ce-Ni catalyst