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Extended Abstract

Ni/SBA-15 catalysts for methane dry reforming

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Abstract

Ten catalyst samples, with increasing nickel loadings of 4 to 29 wt. %, were prepared by impregnation of different nickel precursors (acetate, nitrate, and citrate) in different concentrations on SBA-15 mesoporous silica support. A modified sol-gel method was used to prepare the support, which proved to have a high BET surface area of 834 m2/g, a very well-ordered mesoporous structure and narrow pore size distribution around 7.8 nm. An exhaustive physical, chemical and dynamic characterisation of the prepared samples was performed and an optimised catalyst with optimum activity was proposed. XRD, SEM/EDS, XPS, TEM, and N2 adsorption isotherm were used to assess the bulk and surface structure, morphology and texture of the prepared samples. Good dispersion and reasonably high loadings were obtained from less concentrated solutions of nickel acetate precursors, while the highest loading is obtained using nickel nitrate as precursor. The dry reforming of methane (DRM) reaction, using a model biogas, CH4/CO2 ratio of 1.5:1, was operated isothermally at 550, 600, 650 and 700°C, respectively. The carbon deposition on the catalyst surface was surveyed by XRD, SEM, and TGA/DSC and TPO measurements. There was no carbon formed over the ex- NI acetate samples while for the ones obtained from Ni-nitrate, the amount of carbon increased with the Ni loading. decomposition and Boudouard reaction. Therefore, many research studies were focused on the preparation of supported catalysts with improved activity and stability, by using different preparation methods and different supports. This work deals with the preparation of newly-developed nickel catalysts supported on mesoporous SBA-15 silica. A modified sol-gel method was chosen for the support preparation and impregnation method was used to incorporate the active phase, namely nickel, onto the support. The prepared Ni/SBA-15 catalysts with different Ni loading and different Ni precursor were tested in DRM reaction. The rapid increase in emissions of major greenhouse gases such as CO2 and CH4 in the last decade has seriously affected climate change and the living environment in the world. Besides, syngas, the variable mixture of carbon monoxide and hydrogen, is an important intermediate for downstream processes in the chemical industry. Including many benefits for producing syngas from available natural gas reservoirs, along with the consumption of two main greenhouse gases, bireforming of CH4 using CO2 and H2O (reaction (1)) is the most suitable method with extensive application in the chemical industry The bireforming process (BRM) (reaction (1)) consists of two reactions: dry reforming (DRM) (reaction (2)) and steam reforming (SRM) (reaction (3)): As can be seen from reactions (1) and (2), the enthalpy at 298 K of DRM is +247.3 kJ/mol, 1.122 times higher than BRM. This means that, as an endothermic reaction, the combined CO2 and steam reforming of methane (Equation (1)) consumes less energy than DRM (Equation (2)). Furthermore, this combination shows the most desirable stoichiometry of H2 and CO, completely suitable for the Fischer-Tropsch synthesis process. By feeding CO2 and steam simultaneously, the products' ratio can be controlled by adjusting the CH4/CO2/H2O ratio of the feedstock. Moreover, one of the disadvantages of steam reforming methane is the generation of large amounts of CO2 in the side reaction In our previous study nanosized NiO/SBA-15 catalysts with NiO crystallite size in the range of 12.9 to 18.3 nm were successfully prepared. In this catalytic system, there are 5–6 nm NiO particles dispersed inside the pores and the NiO particles of 20–50 nm distributed on the surface of SBA-15 when Ni content was 23.5–39.2 wt.%. Dispersion of metallic sites into the pores could prevent Ni from sintering and metal loss during reaction. The high dispersion of NiO in the Ni/SBA-15 catalyst is caused by the unique properties of SBA-15 such as uniform pores with large diameters (5.3–6.0 nm), high porosity, and high specific surface area (613 m2/g). The reduced catalysts have high activity in bireforming reaction, reaching 86% CH4 and 77% CO2 converted at 700°C or 90.5% and 80%, respectively, at 800°C. The catalysts work stably for hundreds of hours due to the presence of weak and strong Lewis basic sites which limit coke formation and increase CO2 adsorption. Similarly, Zhang et al. Danilova et al. suggested that the structural similarity of NiO and MgO led to the formation of a solid solution in the form of a thin layer, surrounding the Ni particles that increased catalyst stability and reduced carbon deposition. Wang et al. demonstrated that in the MgO-modified Ni/SBA-15 catalyst synthesized by the coimpregnation method, the MgO particles covered the walls of channels of SBA-15. Besides, the catalyst basicity increased and the Ni dispersion was improved, leading to an enhancement in chemical adsorption of CO2. As a result, the activity and the coke resistance of the catalyst in the dry reforming reaction were improved. In addition, Alipour et al.

However, to the best of our knowledge, the impact of treatment with aqueous NH3 step in catalyst preparation or promoting catalysts by MgO on the activity of Ni/SBA-15 catalyst in bireforming has not been carried out so far. In our previous investigation, it was reported that NiO/SBA-15 catalyst contained 31.4 mass% Ni (Ni/SBA-15), exhibiting higher activity than other Ni/SBA-15 samples. The conversion of CH4 and CO2 on this catalyst in bireforming (BRM) reached 90% and 76%, respectively, at 700°C. In this study, the effect of NH3 alkalization or promotion by MgO on catalytic activity of Ni/SBA-15 catalyst in BRM reaction was investigated and

optimized

Biography:

Dr. Rawaz Ahmed: She worked at Teesside University, with valuable experience in heterogeneous catalysis and sustainable technology having won the top poster prize in catalysis by Royal Chemical Society at Johnson Matthey Conference, Biillingham in March, 2011, and the optimized preparation method and results obtained over the catalyst with optimal formulation are the subject of a patent application (P136962GB; New UK Patent Application; Supported Metal Catalyst; Teesside University). She worked as Research Associate: biomass carbonization at Teesside University after her PhD study

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