O12 : Synthesis gas production from dry reforming of methane over Ni spinel catalysts

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Abstract :

The reforming of methane to syngas using CO_2 instead of steam is an attractive route, including from the point of view of sustainability because it uses two greenhouse gases. Compared to noble metals, transition metals and in particular nickel catalysts are cheap but very easily deactivated by coking. Ni-based mixed oxides structurally well-defined like perovskites and spinels are being studied because they possibly make solid solutions and allow varying the composition and thus the catalytic properties [1]. In this work, the nickel ferrite catalysts were synthesized by coprecipitation with ammonia (CP), hydrothermal synthesis at 140 °C for 12 h (HT) [2] and sol-gel method using polyacrylic acid (SG). Samples were characterized by B.E.T, XRD at room temperature (RTXRD) and high temperature (HTXRD), Raman spectroscopy, SEM-EDX, TEM, TPR and XPS. XRD patterns of all synthesized oxides showed the presence of $NiFe_2O_4$ inverse spinel, confirmed by Raman spectroscopy by the main lines at 488(m), 658(sh), 702(s) cm^{-1}. Hematite (227 and 292 cm^{-1}) was present only in CP case. Depending on the synthesis method, the surface area (B.E.T.), particle size (Rietveld refinement), as well as the surface Ni/Fe atomic ratio (XPS) and the behavior upon reduction varied. According to EDS and XPS, the surface Fe/Ni is close to stoichiometry except for CP. HTXRD in 3%H_2/N_2 up to 800 °C showed that the lines of the spinel vanished above ca. 450 °C. Metallic nickel appeared up to 580 °C and the final phase was Fe-Ni alloy. The catalytic conversion of methane and of CO_2 is very high in the SG case (more 85 % of conversion). A significant contribution of reverse water gas shift reaction explained the low values of H_2/CO ratio. Correlations between preparation method and textural, structural and reducibility of catalysts with catalytic performance will be presented.

References:

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