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Ni Mixed Oxides Type-Spinel for Energy: Application in Dry Reforming of Methane for Syngas (H2 and CO) Production

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Abstract : In the recent years, the dry reforming of methane has received considerable attention from an environmental view point because it consumes and eliminates two gases (CH4 and CO2) responsible for global warming by greenhouse effect. Many catalysts containing noble metal (Rh, Ru, Pd, Pt and Ir) or transition metal (Ni, Co and Fe) have been reported to be active in this reaction. Compared to noble metals, Ni-materials 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 to vary the composition and thus the performances properties. In this work, nano-sized nickel ferrite oxides are synthesized using three different methods: Co-precipitation (CP), hydrothermal (HT) and sol gel (SG) methods and characterized by XRD, Raman, XPS, BET, TPR, SEM-EDX and TEM-EDX. XRD patterns of all synthesized oxides showed the presence of NiFe2O4 spinel, confirmed by Raman spectroscopy. Hematite was present only in CP sample. Depending on the synthesis method, the surface area, particle size, as well as the surface Ni/Fe atomic ratio (XPS) and the behavior upon reduction varied. The materials were tested in methane dry reforming with CO2 at 1 atm and 650-800 °C. The catalytic activity of the spinel samples was not very high (XCH4 = 5-20 mol% and XCO2 = 25-40 mol %) when no pre-reduction step was carried out. A significant contribution of RWGS explained the low values of H2/CO ratio obtained. The reoxidation step of the catalyst carried out after reaction showed little amounts of coke deposition. The reducing pretreatment was particularly efficient in the case of SG (XCH4 = 80 mol% and XCO2 = 92 mol%, at 800 °C), with H2/CO > 1. In conclusion, the influence of preparation was strong for most samples and the catalytic behavior could be interpreted by considering the distribution of cations among octahedral (Oh) and tetrahedral (Td) sites as in (Ni2+1-xFe3+x) Td (Ni2+xFe3+2-x) OhO2-4 influenced the reducibility of materials and thus their catalytic performance.

Keywords : NiFe2O4, dry reforming of methane, spinel oxide, oxide zenc
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