Poster Presentation

MS06.P01

XANES studies of zirconia-ceria/Ni during partial/total methane oxidation

R. Bacani[1], M. Fantini[1], T. Martins[2], S. Larrondo[3], D. Lamas[4]

[1]Universidade de São Paulo, Departamento de Física Aplicada, São Paulo, Brazil, [2]Universidade Federal de São Paulo, Instituto de Ciências Ambientais, Químicas e Farmacêuticas, Diadema, Brasil, [3]Centro de Investigaciones en Sólidos, CINSO-UNIDEF-CONICET, Buenos Aires, Argentina, [4]Universidad Nacional del Comahue, Laboratório de Caracterización de Materiales, Facultad de Ingeniería, Neuquén, Argentina

Zirconia-ceria solid-solutions are extensively used as promoters for three-way catalysts, in addition, these materials can be used as anodes in solid oxide fuel cells (SOFCs) operated with hydrocarbons. The structural features of ZrO2-CeO2 materials in combination with oxygen storage/release capacity (OSC) are crucial for various catalytic reactions. The direct use of hydrocarbons as fuel for the SOFC (instead of pure H2), without the necessity of reforming and purification reactors can improve global efficiency of the system. The samples preparation method was developed using Zr and Ce chloride precursors, HCl aqueous solution, Pluronic P123, NH4OH and a Teflon autoclave. The samples were dried and calcined, until 540ºC. The NiO impregnation was made with an ethanol dispersion of Ni(NO3)×6H2O, calcinated in air until 350ºC for 2 hours. In-situ XANES experiments are capable to evaluate the reduction/oxidation potencial of Ni and Ce species in ZrO2-CeO2/Ni samples during partial/total methane oxidation and reduction reactions with H2. The experiments at the Ni K-edge/Ce L3-edge were collected at the LNLS D06A-DXAS beam line in transmission mode, using a Si(111) monochromator and a CCD camera as detector. The data were acquired during a series of temperature programmed reduction steps (TPR), under a 5% H2/He until 600ºC, and mixtures of 20%CH4:5%O2/He with 2:1, 1:1 and 1:2 ratios. After each process with CH4 and O2, a TPR procedure was performed in order to evaluate the reduction capacity of the sample after reactions with CH4. The results demonstrated that NiO embedded in the porous ZrO2-CeO2 matrix, reduces at lower temperatures than standard NiO, measured in the same conditions, revealing that the mesoporous support improves the reduction of impregnated NiO. For both edges, there was formation of H2 during partial methane oxidation at 600ºC. The total oxidation of methane was observed in lower temperatures (500ºC). These results reveal that a high ceria content (90%) could be a great candidate for the SOFC anode.

[1] *M. Mamak, N. Coobs, G. Ozin, et al., Chem. Soc., 2000, 122, 8932-8939,* ***[2]*** *J. Kaspar, P. Fornasiero, M. Graziani, Catal. Today 1999, 12, 285-298,* ***[3]*** *S. Larrondo, A. F. Craievich, D. G. Lamas et al, Catal. Today, 1999, 107-108, 53-59*

Keywords: XANES, catalisys, zirconia-ceria