Hydrogen Production from Auto-Thermal Reforming of Ethanol Catalyzed by Tri-Metallic Catalyst

Authors : Patrizia Frontera, Anastasia Macario, Sebastiano Candamano, Fortunato Crea, Pierluigi Antonucci Abstract : The increasing of the world energy demand makes today biomass an attractive energy source, based on the minimizing of CO2 emission and on the global warming reduction purposes. Recently, COP-21, the international meeting on global climate change, defined the roadmap for sustainable worldwide development, based on low-carbon containing fuel. Hydrogen is an energy vector able to substitute the conventional fuels from petroleum. Ethanol for hydrogen production represents a valid alternative to the fossil sources due to its low toxicity, low production costs, high biodegradability, high H2 content and renewability. Ethanol conversion to generate hydrogen by a combination of partial oxidation and steam reforming reactions is generally called auto-thermal reforming (ATR). The ATR process is advantageous due to the low energy requirements and to the reduced carbonaceous deposits formation. Catalyst plays a pivotal role in the ATR process, especially towards the process selectivity and the carbonaceous deposits formation. Bimetallic or trimetallic catalysts, as well as catalysts with doped-promoters supports, may exhibit high activity, selectivity and deactivation resistance with respect to the corresponding monometallic ones. In this work, NiMoCo/GDC, NiMoCu/GDC and NiMoRe/GDC (where GDC is Gadolinia Doped Ceria support and the metal composition is 60:30:10 for all catalyst) have been prepared by impregnation method. The support, Gadolinia 0.2 Doped Ceria 0.8, was impregnated by metal precursors solubilized in aqueous ethanol solution (50%) at room temperature for 6 hours. After this, the catalysts were dried at 100°C for 8 hours and, subsequently, calcined at 600°C in order to have the metal oxides. Finally, active catalysts were obtained by reduction procedure (H2 atmosphere at 500°C for 6 hours). All sample were characterized by different analytical techniques (XRD, SEM-EDX, XPS, CHNS, H2-TPR and Raman Spectorscopy). Catalytic experiments (auto-thermal reforming of ethanol) were carried out in the temperature range 500-800°C under atmospheric pressure, using a continuous fixed-bed microreactor. Effluent gases from the reactor were analyzed by two Varian CP4900 chromarographs with a TCD detector. The analytical investigation focused on the preventing of the coke deposition, the metals sintering effect and the sulfur poisoning. Hydrogen productivity, ethanol conversion and products distribution were measured and analyzed. At 600°C, all tri-metallic catalysts show the best performance: H2 + CO reaching almost the 77 vol.% in the final gases. While NiMoCo/GDC catalyst shows the best selectivity to hydrogen whit respect to the other tri-metallic catalysts (41 vol.% at 600°C). On the other hand, NiMoCu/GDC and NiMoRe/GDC demonstrated high sulfur poisoning resistance (up to 200 cc/min) with respect to the NiMoCo/GDC catalyst. The correlation among catalytic results and surface properties of the catalysts will be discussed.

Keywords : catalysts, ceria, ethanol, gadolinia, hydrogen, Nickel

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