FREE ARTICLE

$La_{0.5}Ce_{0.5}O_{1.75}$-Catalytic Layer for Methane Conversion into C_2 Products Using Solid Oxide Fuel Cell

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Abstract

Methane (CH_4), the major constituent of natural gas and biogas, is an abundant source to obtain value-added hydrocarbons. The oxidative coupling of methane (OCM) is a direct catalytic route to convert CH_4 towards C_2 hydrocarbons, ethane (C_2H_6) and ethylene (C_2H_4). Using a solid oxide fuel cell (SOFC) is a strategy to overcome some challenges of fixed-bed catalytic reactors. In this context, we have studied the $La_{0.5}Ce_{0.5}O_{1.75}$ (LCO) oxide as a catalytic layer in a SOFC for methane conversion to ethylene. Single phase LCO was synthesized by the combustion method. X-ray diffraction (XRD) showed that LCO solid solution has a disordered fluorite crystalline structure. Raman spectroscopy data evidenced the presence of surface oxygen vacancies, which may benefit the OCM reaction. SEM images evidenced a microstructure composed of porous agglomerated particles with an irregular shape, expected from the combustion synthesis. Impedance spectroscopy (IS) measurements of sintered LCO pellets were performed in a wide range of both temperature and oxygen partial pressure (pO_2). Thermally activated behavior of the total electrical conductivity revealed that LCO is predominantly an ionic conductor at $10^{-6} < pO_2 < 0.21$ atm, whereas at $pO_2 \sim 10^{-21}$ atm, n-type electronic conductivity leads to a mixed ionic electronic conductor behavior, due to the $Ce^{4+/3+}$ reduction. The catalytic properties of LCO and the optimized OCM reaction conditions were investigated a fixed-bed reactor. The desired products were obtained (C_2H_6 and C_2H_4), as well the parallel products (CO_x, and traces of C_3H_8 and C_4H_{10}). The higher rates of CH_4 conversion (22 %), C_2 selectivity (54 %) and yield (12 %) were reached for the reaction developed with $4CH_4$:$1O_2$ at 750 ºC. Electrolyte-supported single cells were prepared using a YSZ disk (diameter ~ 19 mm, thickness ~ 0.4 mm) as the electrolyte. The cathode (LSM) and anode (NiO/YSZ) layers were both deposited by screen-printing on each side of the YSZ disk. For the catalytic layer, LCO was deposited, using the spray-coating technique, on the anodic side. The electrochemical properties of such fuel cell tests were characterized under 10 % H_2 between 750 – 800 ºC, and then, the fuel was switched to 10 % CH_4, both using Helium as a carrier gas. The anode outlet gas was monitored by an online gas analyzer setup that showed formation of C_2H_4, C_2H_6 and CO_x, indicating the effectiveness of the catalytic layer.

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