

Ni–Ru-containing mixed oxide-based composites as precursors for ethanol steam reforming catalysts: effect of the synthesis methods on the structural and catalytic properties

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To date, hydrogen is the most environmentally friendly fuel for various energy and heat generators (fuel cells, internal combustion engines and mobile power plants). In context of the green energy of the future, hydrogen is associated with a promising technology of electrochemical generators based on solid oxide fuel cells (SOFCs) with internal or external reformer of fuels, whose main qualities are environmental friendliness, mobility and high efficiency. Moreover, due to fuel source flexibility, such devices can successfully provide reforming of various carbon-containing fuels, including bio-renewable ones. Among others, a great attention is paid to the ethanol steam reforming process. Since the catalyst for internal fuel reforming in SOFC is a multifunctional layer supported on the anode, it must satisfy many requirements: (1) activity in the reaction of steam reforming of oxygenates (breaking C–C and C–H bonds), (2) thermochemical stability (resistance to sintering and carbonization), (3) compatibility with anode layers (stability to delamination and cracking) and (4) mixed ionic/electron conductivity. Today, it is known that some nanocomposite materials, including complex oxides of transition and rare-earth elements (RRE) with the structures of perovskite, fluorite, or spinel, have the aforementioned properties. Today, the synthesis of such composites with optimal characteristics is an urgent task of hydrogen energy. In this study, catalyst precursors based on perovskite–fluorite nanocomposites with the general formula $[\text{LaMn}_{1-x}\text{B}_x\text{O}_{3+\delta}/\text{Ln}_{1-y}\text{Zr}_y\text{O}_2]$ (1:1 by mass), B = Ni, Ru, Ln = Pr, Sm, Ce were synthesized by three different methods. Two synthesis methods — sequential polymeric method (formation of Ni- and Ru-containing perovskite from a polymer matrix in the presence of already formed fluorite oxide) and ultrasonic dispersion of as-prepared complex oxides in isopropanol with addition of surfactant — provide formation of nanocomposites with a developed interphase between phases. In the one-pot synthesis method from a polymer containing all cations, the perovskite phase is not formed due to La incorporation into the fluorite matrix, with inclusions of amorphous Mn oxides and NiO phase being present. As the result, the highest concentration of surface metal sites in reduced catalysts was revealed for nanocomposite prepared by the sequential polymeric method, while the lowest concentration was observed for samples prepared by the one-pot method. Catalytic activity of nanocomposites in ethanol steam reforming correlates with the surface density of metal sites. They are stable to coking and provide full conversion of ethanol and hydrogen yield above 70% at 650°C for 10 h.