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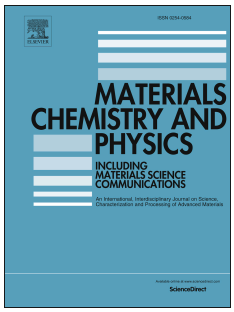

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Sulfated $Ce_xZr_{1-x}O_2$ oxides. Surface properties and performance for methane oxidation under fuel-rich conditions

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Abstract

Two series of sulfated zirconia-ceria based catalysts have been prepared by coprecipitation of the Zr^{4+} and Ce^{3+} salts, which were then sulfated by impregnation using H_2SO_4 and $[(NH_4)_2S_2O_8]$ and calcined at 650 ºC for 4 h. The sulfated and persulfated Zr-Ce-O samples were synthesized with Zr/Ce ratios ranging from 1 to 15. The textural, structural and surface characteristics of the resulting modified catalysts were determined by N_2 adsorption-desorption, XRD, XPS spectroscopy and TPD of NH_3, and results were related to their catalytic behaviour. The catalysts were tested in the direct conversion of methane at high temperature (650 ºC) under fuel-rich conditions to obtain C_1 oxygenates (CH_3OH and HCHO). Experiments were conducted using low O_2/CH_4 ratio mixtures and high space velocity. It was shown that O_2/CH_4 feeds resulted in practically total combustion of CH_4 when sulfated/persulfated ceria-zirconia catalysts are used, while the nature of the different phases modifies the surface acidity, which plays a crucial role on the extent of deep oxidation reactions.

Keywords: ceria-zirconia mixed oxides, acidity, surface characterization, partial oxidation of methane.

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