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Partial oxidation of methane on Pt-supported lanthanide doped ceria–zirconia oxides: Effect of the surface/lattice oxygen mobility on catalytic performance

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ABSTRACT

Partial oxidation of methane into syngas at short contact times (5–15 ms) was studied in both steady-state and transient modes at temperatures up to 850 °C in realistic feeds (CH_4 content up to 20%, $CH_4/O_2 = 2$) with a minimum impact of mass and heat transfer for structured catalysts carrying $Pt/Ln_{0.3}Ce_{0.35}Zr_{0.35}O_{2-y}$ (Ln = La, Pr, Gd) as thin layers on walls of corundum channel substrates. Oxygen mobility and reactivity of the active phase were characterized by oxygen isotope heteroexchange, temperature-programmed O_2 desorption and CH_4 reduction, isothermal pulse reduction by methane with wide variation of CH_4 concentrations and TAP pulse studies. Experimental data point towards a selective oxidation of methane into syngas via a direct route with oxygen-assisted methane activation. This mechanistic feature is related to the strong Pt-support interaction stabilizing highly dispersed oxidic Pt species less active in CH_4 and syngas combustion than metallic Pt clusters. Support activates O_2 molecules and supplies active oxygen species to Pt sites. A high rate of oxygen diffusion on the surface and in the bulk of the support and Pt-support oxygen spillover stabilizes Pt in a well dispersed partially oxidized state while preventing coking at high concentrations of CH_4 in the feed.