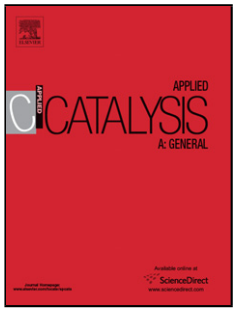

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Surface chemistry and reactivity of $Pd/BaCeO_3 \cdot 2ZrO_2$ catalyst upon sulphur hydrothermal treatment for the total oxidation of methane

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Highlights

► 2% $Pd/BaCeO_3 \cdot 2ZrO_2$ catalyst prepared by solution combustion synthesis ► Exposed up to 450 h sulphur hydro-thermal ageing treatment ► Intermediate better performance due to ZrO_2 and CeO_2 segregation acting as O_2 donor ► Final worsening of the performance towards methane combustion ► Ageing mechanisms: subsurface-bulk sulphates formation and oxidation of surface Pd^0

ABSTRACT

Starting from metal nitrates and glycine, 2% $Pd/BaCeO_3 \cdot 2ZrO_2$ catalyst was prepared by solution combustion synthesis and tested towards the total oxidation of methane. The catalyst underwent heavy sulphur-hydrothermal treatment at 800 °C up to 450 h. The catalyst was fully characterized (XRD, BET, SEM, O_2-TPD, FTIR analysis and catalytic activity via CH_4-TPC) every 150 h. With ageing, catalytic activity tests demonstrated that the catalyst was heavily poisoned after 150 h; then it recovered the catalytic activity after 300 h, with a resulting performance better than the one reached in the fresh status. At the end of the sulphur-hydrothermal treatment, after 450 h, the catalyst resulted heavily poisoned again. On the fresh catalyst surface, IR analysis of CO adsorption evidenced the formation of highly dispersed Pd metal clusters and Pd ions. After 300 h of sulphur hydrothermal ageing, the increased catalytic activity towards methane combustion was probably supported by the segregation of ZrO_2 and CeO_2. Moreover, subsurface and bulk sulphate formation was detected with ageing and Pd metal species were not anymore available for CO coordination, probably because hindered by sulphate deposits. Prevailing ageing mechanisms resulted in the