Synergetic and precursor effects in propane total oxidation over Cu- and Ce-based catalysts

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Introduction

Volatile organic compounds (VOC’s) exhibit a significantly high vapour pressure at ambient conditions. These VOC’s are responsible for increased levels of ozone in the troposphere and for ozone depletion in the stratosphere. According to Theloke and Friedrich, one third of all emitted VOC’s are alkanes [1]. Emissions from both stationary and non-stationary sources are subjected to an ever stricter legislation, due to an increasing environmental awareness. Catalytic oxidation offers the advantage over thermal incineration that these VOC’s can be removed from effluent streams in low concentrations and at relatively low temperatures [2]. This paper describes the total oxidation of propane as model reaction over Cu- and Ce-based catalysts by means of steady-state experiments.

Acquisition of experimental data

Four catalysts are used: a CeO-CeO₂/γ-Al₂O₃ catalyst, formally denoted as CuCeN, synthesized by impregnation of a γ-Al₂O₃ carrier with precursors Cu(NO₃)₂ and Ce(NO₃)₃; catalysts Cu-N and Cu-C are made by impregnation of θ-Al₂O₃ with Cu(NO₃)₂ and CuCO₃; a Ce-N catalyst is obtained by impregnation of a γ-Al₂O₃ carrier with Ce(NO₃)₃. These four catalysts are tested in the total oxidation of propane using a conventional plug flow reactor with a propane and oxygen inlet partial pressure of 0.58 kPa and 3.51 kPa at three temperatures: 595, 621 and 648 K. For sample Ce-N, an extra temperature level of 634 K is applied. The space time is varied up to 570 kgₑₜₐₜₙ s mol⁻¹.

Results

The experimental data are regressed by a Mars-van Krevelen (MVK) model. This model corresponds to a reduction step in which propane reacts with a single oxidized site and with a reoxidation step by gas phase oxygen, involving a single reduced surface site.

Conclusions

A CuO-CeO₂/γ-Al₂O₃ catalyst synthesized by impregnation with nitrate precursors is the best catalyst for the oxidation of propane from a series of four copper oxide and ceria-based catalysts. Whereas the reduction step seems to be the most critical step in the reaction mechanism on Cu-based catalysts, it is the reoxidation that is critical on a CeO₂ based catalyst. A mixed oxide CuO-CeO₂/γ-Al₂O₃ allows combining the good characteristics of both types of oxides in a synergetic way. A precursor effect is observed by using two copper oxide catalysts, made by impregnation of Cu(NO₃)₂ and CuCO₃ on θ-Al₂O₃. The latter catalyst shows higher propane conversion as well as TOF.

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