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Kinetics and mechanism of the deep oxidation of propane over Pt-fiberglass catalyst

BS Bal'zhinimaev

Boreskov Institute of Catalysis, Russia

A detailed kinetic study on the deep oxidation of propane was carried out in a wide range of temperatures and concentrations using a novel Pt-fiberglass catalyst. Highly dispersed 1nm Pt²⁺-Pt⁰ metal oxide clusters are confined in the bulk of glass at a depth up to 20nm. In spite of extremely low platinum content (0.01-0.02%) this catalyst showed a very high activity in the propane oxidation. The turnover frequency (TOF) value is by 1-2 orders of magnitude higher than the turnover frequencies of Pt catalysts with conventional metal oxide supports due to capacity to activate molecular oxygen. The modified Langmuir-Hinshelwood mechanism, where the key step is based on the ability of active Pt²⁺-Pt⁰ sites to simultaneously adsorb molecular oxygen and propane, was used to propose the kinetic model of the reaction that adequately describes such experimental features like the extremal dependence of the reaction rate on the O₂/C₃H₈ ratio, the sharp changes of reaction order with respect to oxygen, the significant growth of apparent activation energy with decreasing the oxygen concentration, etc. In particular, the extremal dependence is caused by competitive adsorption of propane and oxygen. At high ratios, the adsorbed oxygen occupies virtually all active sites that are accessible for propane adsorption, while in the case of oxygen deficiency, there occurs irreversible adsorption of propane with coke formation. In both cases, the reaction rate and reaction order sharply decrease. The maximum reaction rate is attained at the optimal O₂/C₃H₈ ratio when the oxidation involves the maximum number of active sites.

balzh@catalysis.ru