Laser diagnostics and kinetic modelling of reaction intermediates in catalytic combustion

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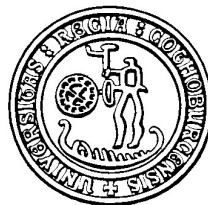
Laser diagnostics and kinetic modelling of reaction intermediates in catalytic combustion

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Abstract

Catalytic combustion of hydrogen has been studied on hot polycrystalline palladium and platinum catalysts using laser spectroscopy. The OH radical, which is an important intermediate in water formation, was probed outside the catalysts using laser–induced fluorescence (LIF) and cavity ringdown spectroscopy (CRDS). To complement the experiments, kinetic models were also derived using the CHEMKIN simulation package. The experiments and models were performed in a stagnation–point flow field geometry. The OH desorption and water production were measured outside Pd as a function of the hydrogen mixing ratio, α_{H2}, at a temperature of 1300 K, pressures between 13–26 Pa and flows between 100–200 $SCCM$ using LIF and microcalorimetry. The yield of OH had a maximum at α_{H2}=10% while the maximum in water production occurred at α_{H2}=40%. The apparent desorption energy of OH outside a Pd catalyst was also measured with LIF as a function of α_{H2}. From kinetic modelling the OH desorption energy on Pd was found to have a first–order coverage dependence according to: $E_{OH}^d(\theta) = E_{OH}^d(0) - B\theta$, where $E_{OH}^d(0)$ was the desorption energy at zero coverage, B a constant and θ the total coverage. The desorption energy at zero coverage, $E_{OH}^d(0)$, was determined as 226 kJ/mol and the coverage–dependent desorption energy $E_{OH}^d(\theta)$ was calculated as a function of α_{H2}. The coverage on Pd was also derived as a function of α_{H2}. The hydrogen addition reaction, $H + OH \rightleftharpoons H_2O$, was determined to be the main route of water formation on Pd at 1300 K. Exact number densities of short–lived intermediates are valuable to improve theoretical models. In this work OH radicals outside a polycrystalline Pt catalyst have for the first time been quantified using cavity ringdown spectroscopy.

Keywords: Catalysis, Combustion, Kinetic modelling, Palladium, Platinum, Water formation, OH hydroxyl, Laser–induced fluorescence, Cavity ringdown spectroscopy, Desorption energy, CHEMKIN.