344d Direct Vs Indirect Oxidation Pathways in Autothermal Catalytic Partial Oxidation of Methane over Pt

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Catalytic partial oxidation of methane (CPOM) at high-temperature, millisecond contact-times (T > 900oC, □ = 1-50 ms) is a technologically interesting reaction pathway for the production of synthesis gas or hydrogen. While many studies on CPOM over noble metal catalysts have been published in the past ten years, much debate exists in the scientific community whether synthesis gas is produced via indirect or direct pathways. Unfortunately, the extreme reaction conditions make conventional kinetic studies as well as surface science investigations of the reaction mechanism virtually impossible. Here, we report results from a study into the reaction mechanism of CPOM over Pt catalysts. Catalyst contact times were varied and product concentrations were correlated with in-situ temperature profiles measured throughout the catalyst bed. The results give convincing evidence that the reaction proceeds in two stages: initial direct oxidation to CO and H2O (at □ < 2 ms) followed by steam reforming of methane. Water gas shift plays only a minor role at all experimental conditions. H2 is produced predominantly via indirect oxidation while CO is produced predominantly via direct oxidation. The results indicate that the distinction between “direct” and “indirect” formation of synthesis gas is meaningless and need to be answered separately for the carbon and hydrogen pathways.