LSM-Webinar

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Homogeneous ignition of H2/CO/O2/N2 mixtures over palladium at pressures up to 8 bar

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Abstract:
The catalytic and gas-phase combustion of fuel-lean $H_2/CO/O_2/N_2$ mixtures over palladium was investigated experimentally and numerically at a global equivalence ratio $\varphi = 0.285$, H_2:CO volumetric ratios 1-4, pressures 1-8 bar and catalyst surface temperatures 950-1200 K. In situ planar laser induced fluorescence (PLIF) of the OH radical monitored homogeneous combustion inside a channel-flow catalytic reactor, while 1-D Raman measurements of main gas-phase species concentrations across the channel boundary layer assessed the heterogeneous processes. Simulations were carried out with a 2-D numerical code using detailed heterogeneous and homogeneous chemical reaction mechanisms and realistic transport. The simulated and measured transverse species profiles attested to a transport-limited catalytic conversion of H_2 and CO at all operating conditions. The OH-PLIF measurements and the simulations confirmed the establishment of appreciable homogeneous combustion only for $p < 4$ bar, with progressively diminishing gas-phase contribution as the pressure increased from 4 to 8 bar. This strong pressure dependence reflected the complex pressure/temperature dependence of the homogeneous ignition chemistry as well as the competition between the catalytic and gaseous reaction pathways for H_2 and CO consumption. Over the gaseous induction zones ($x < x_{ig}$), the wall temperatures were below the pressure-dependent upper temperature limit for the decomposition of PdO to metallic Pd. Even though palladium catalysts exhibited a “self-regulating” temperature effect due to the decomposition of PdO, the attained temperatures were still sufficient to ignite homogeneous combustion of the $H_2/CO/O_2/N_2$ mixtures, in contrast to hydrocarbon fuels for which gas-phase combustion was largely suppressed over PdO in the pressure range 1-8 bar. The results indicated that for the elevated pressures and preheats of syngas-fueled hetero-/homogeneous combustion power systems, gas-phase chemistry cannot be ignored during reactor design.

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