New insights from studies of Pd single crystals during CO oxidation in a stagnation flow

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In-situ knowledge of the gas phase around a catalyst is essential to make accurate correlations between catalytic activity and surface structures in operando studies [1]. Planar laser-induced fluorescence (PLIF) has been shown as a powerful gas detection technique which can image the gas distribution in the vicinity of a catalytic sample with high spatial and temporal resolution, and our previous studies have shown that the gas partial pressures close to the sample measured by PLIF differ significantly from those measured by a conventional mass spectrometer connected to the gas outlet of a reactor [2,3].

In this work, we have studied the CO oxidation over a Pd(100) single crystal in a stagnation flow reactor through visualizing the CO₂ distribution over the sample by PLIF, and compared the experimental results with theoretical modelling [4]. We found that with stagnation flow geometry, we can get well-defined CO₂ signal from the surface of the sample, i.e. not interfered by the signal generated from other parts of sample (e.g. sides), as clearly visualized by PLIF both spatially and temporally. This indicates that from experimental point of view, the stagnation flow geometry has a distinct advantage over other flow geometries for operando studies with single crystals. In addition, we have obtained a very good match between our experimental and modelling data in characterizing the stagnation flow. This indicates a great potential of using the measured turn over frequency (TOF) over the sample surface by PLIF, to simulate the chemistry of a catalyst by theory.

Figure 1. Measured CO₂ distribution over a highly active Pd(100) single crystal during CO oxidation in a stagnation flow

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