Laser diagnostics for catalytic reactions

Background: Catalytic systems are widely used to reduce emissions of greenhouse gases and local pollutants in the chemical, energy and transportation sectors. The system is dynamic. The catalyst changes its activity in space and time due to spatial and temporal variations of the fluid phase and the time of operation. The gas-phase species concentration over the catalytic surface is often probed with thin capillaries. The invasiveness of such sampling techniques is challenging for the quantitative interpretation of the measurement results. Moreover, the results are often limited to point data or 1D profiles in streamwise direction. Therefore, there is an urgent need to use non-invasive methods, such as laser based techniques, to visualize the spatial and temporal development of the gaseous atmosphere over catalyst in operando.



Schematic view on laser diagnostics setup

Project: Our research aims to have a better understanding of the interaction between the catalyst kinetics and the mass transport by obtaining the spatially and temporally resolved reactant/product concentration distribution via laser based techniques, e.g. Planar laser-induced fluorescence (PLIF), Raman spectroscopy. Reactor with good optical access is required for this study. For PLIF measurement, a thin laser sheet is guided into the reactor to excite the target species, e.g., NO, OH, or HCHO. As the laser sheet is oriented parallel to the flow direction and perpendicular to the catalytic surface, 2D images for the distribution of the target species over the catalytic surface could be obtained. Alternatively, the laser can also be used for spectrally resolved 1D-profiles using Raman spectroscopy. The laser is focused into a thin line perpendicular to the catalytic surface, thus the species concentration profiles in wall-normal direction can be obtained. Raman scattering can be used for many major species, e.g., H₂O, HNCO, CH₃OH, CO, CO₂, and other hydrocarbons which are not detectable with LIF. Information of the species concentration gradient perpendicular to the catalytic surface is of great importance. Because in many cases, especially at high temperature or with highly reactive catalyst, the overall reaction rate is not controlled by the intrinsic reaction rate but the mass transfer limitation. Without concentration gradient date in wall-normal direction, the catalytic activity in the transport limited process is very likely to be underestimated. With spatially and temporally resolved laser diagnostics, we can better understand the catalyst behavior in the prescribed flow field.

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