Disentangling Kinetics of CO Oxidation and Mass Transport in Nanoporous Gold using Pulsed Field Gradient NMR

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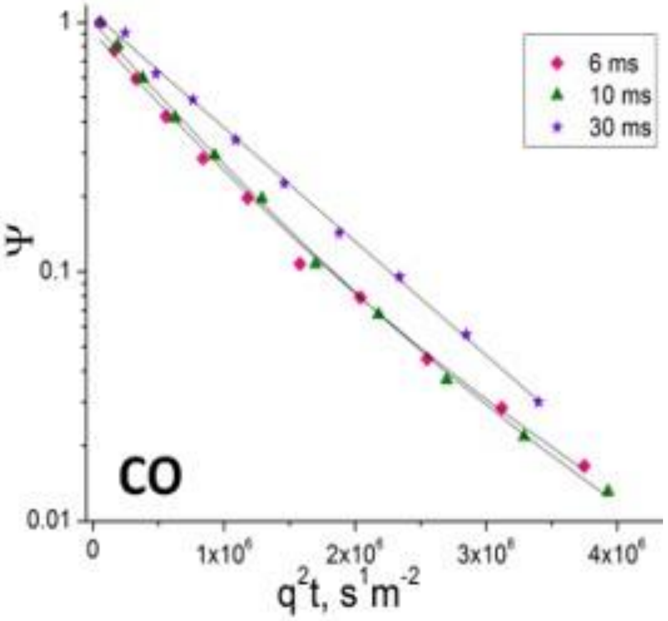
Nanoporous gold, which has pores in the mesopore size range, is a promising candidate for catalytic oxidation of carbon monoxide (CO) to carbon dioxide (CO_2). *A fundamental understanding of molecular diffusion in nanoporous gold is of crucial importance to assess its potential applications for catalysis*.

In this experiment, MagLab users employed pulsed field gradient (PFG) NMR spectroscopy at a high magnetic field (17.6T) to quantify self-diffusion of carbon monoxide and carbon dioxide in beds of nanoporous gold particles. Diffusion measurements were performed via ^{13}C detection over a broad range of diffusion times to measure displacements both smaller and larger than the relevant dimension of the nanoporous gold particles (**see Figure**). *The high magnetic field was needed to achieve sufficiently large signal-to-noise to determine diffusion rates*. For displacements smaller than the particle size (short diffusion times), two populations are resolved for each studied sorbate: (1) molecules that diffuse inside the particles, and (2) molecules that diffuse in the gas phase outside the particles. These observed species have two different diffusivities, and the ratio between these diffusivities is defined as the tortuosity factor.

The tortuosity factor quantifies how much harder it is for gas to diffuse inside the particles compared to outside the particles. Measuring tortuosity factors enables contributions from mass transport to be disentangled from the kinetics of surface reactions (i.e. microkinetics). This high-magnetic-field technique was able to determine the rate constant and turnover frequency for low-temperature CO oxidation without the ambiguities arising in prior measurements from potential transport limitations. *Based on these results, it was possible to predict the optimum catalyst particle size, the size that minimizes the diffusion limitations on the desired reactions, answering the question: which size of nanoporous gold particles is best for use as a catalyst?*



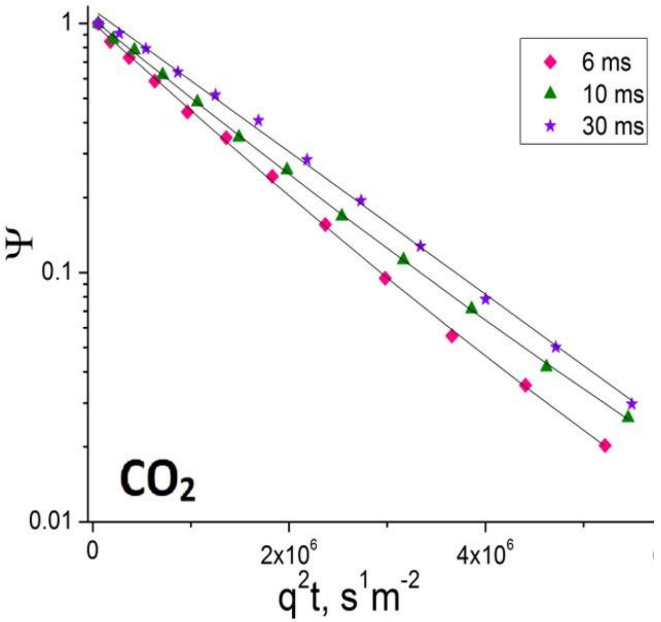


Figure: *^{13}C pulsed-field gradient NMR attenuation curves, measured for gas diffusion inside a bed of nanoporous gold particles that are equilibrated with 15 bar of carbon monoxide (CO, at **left**) or carbon dioxide (CO_2 at **right**). The measurements were performed at 296K for the different diffusion times indicated and at the high magnetic field of 17.6T, which realized the higher signal-to-noise necessary to unambiguously identify the optimum particle size for nanoporous gold to catalytically oxidize carbon monoxide to carbon dioxide.*

Facilities and instrumentation used: AMRIS, 750 MHz/89 mm Bruker Avance III HD

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