Can H/D exchange by neutron imaging anticipate the conversion of hydrogenation catalysts?

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Hydrogenation reactions are crucial in a renewable energy scenario, notably to facilitate the production of hydrocarbons from hydrogen. The reactions are catalyzed by specific catalysts consisting of metal nano-particles on an oxide support. The chemical function of the support is unclear [1]. Hydrogen after dissociative chemisorption may migrate over the oxide surface and thereby promote the transport to and from the reactive sites [2]. Verification of this so-called hydrogen spillover and particular its effect on the reaction kinetic is an analytical challenge, because the quantitative detection of dynamic hydrogen at relatively low concentrations (c << 1 at.%) is difficult.
We developed a combinatorial setup for neutron imaging, capable of measuring up to 69 samples simultaneously under identical conditions (temperature/pressure) to quantify the amount of hydrogen in different hydrogenation catalysts.
For this, hydrogen/deuterium exchange measurements at temperatures between 20 to 350°C and at a pressure of 1bar were performed on Ni deposited on reducible and non-reducible supports. The measurements are the starting point of a universal database of hydrogenation catalysts. To correlate the catalytic activity with H/D exchange observed by neutron imaging, the CO_2 conversion of each catalyst CO_2 as a function of pressure, temperature, and space velocity was measured. The multitude of samples measured simultaneously allowed to analyze the dependence of the activity on specific properties such as metal particle density. This yields additional insights of the generally good correlation between hydrogen-deuterium exchange and catalytic activity.

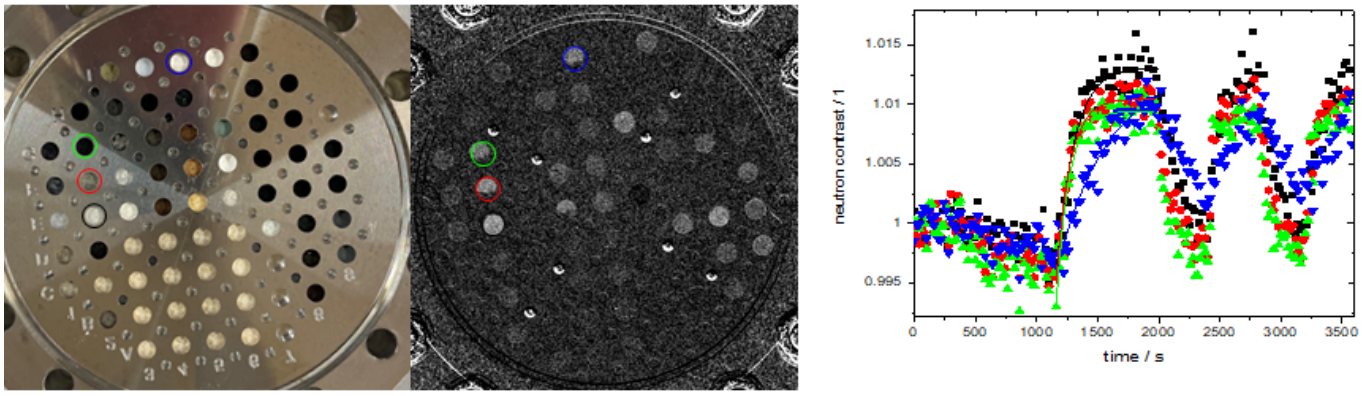

Figure 1: Left: optical micrograph of the combinatorial sample stage. Middle: Neutron image during H-D exchange. Right: Kinetics of H-D exchange as measured by neutron contrast.
Samples: Al O (blue); Ni 0.5%-at on Al O (black); Ni 5%-at on Al2O3 (red); Ni 40%-at on Al O (green)

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