

Neutrons for Catalysis – Summary of *Dynamics* Breakout

Can we probe reactions with neutron scattering?

Requirements for elucidating reaction mechanism (diffusion + conversion)

- Time resolution (ms-s; catalytic cycles) – INS (rapid quench methods)
- Bulk vs. surface – catalysis is surface phenomena
- **Realistic conditions (temperature, pressure) – cell design/compatibility issues**
- Complementary techniques – INS/Raman, VISION spectrometer (simultaneous INS/diffraction), INS-mass spectrometry (*operando*), methods to assess low Z/high Z combinations (Mo₂N)
- Computation – spectra interpretation (INS)

Techniques compatible with goals: **Spectroscopy** (INS) and **Dynamics** (QENS, NSE)

Diffusion

- **Diffusion** of hydrogenous molecules in model systems identified as immediate attainable goal
- Obvious model systems – zeolites (crystalline regularity), pillared clays (2-dimensional diffusion, explore the extent of confinement on diffusional processes)
- Possible outcome – **molecular diffusion mechanism** (jump diffusion) – average jump distance correlates with distance between cations in zeolite, intercalated metals in clays
- H diffusion on surface (spillover) – Role in hydrogenation/dehydrogenation

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Reaction Conversion

- Detection of intermediates by INS: Time-resolution (ms-s) in ***dynamic*** measurements is not there (maybe never) to capture catalytically-cycle intermediate – rapid quench experiments important
- H or other neutron probe molecule (OH, H₂O) to identify adsorption sites/diffusion on M(*hkl*) nanoparticles synthesized by colloidal methods
- Isotopic substitution (partial deuterated/perdeuteration/hydrogen) – isotopic scrambling, isotopomer (Are we in trouble?)

Dynamics in Catalyst Synthesis – completely unexplored!

- Zeolite growth – distinction between internal/external OH, partition coefficient for organocation (SDA) in solid (zeolite precursor) versus solution
- Nanoparticle crystallization, exploit dynamic PDF, complementary to x-ray pdf; selectivity amongst bimetallic constituents

Synthesis with atom-by-atom precision