Electronic Supplementary Material (ESI) for Physical Chemistry Chemical Physics.

Total neutron scattering refers to the measurement and analysis of the complete diffraction pattern including both Bragg and diffuse components. This is a common approach that is frequently applied to systems which exhibit short range order, such as amorphous glasses, liquids or highly disordered crystals. In contrast to X-ray scattering the neutron scattering length and the scattering cross section is not a monotonic function of atomic number, but is atom and isotope dependent. This means that neutron techniques can be used for studies of light elements in the presence of heavy ones or frequently to distinguish structural contributions from elements that are adjacent in the periodic table. Neutron methods are consequently well suited for investigating hydrogenation reactions. The current system is a complex 'disordered system' that combines aspects of liquid-like (reactants and products), glass-like (silica), and semi-crystalline (ordered mesopores) character. A diffraction pattern from highly-crystalline materials is represented by sharp, well-defined peaks corresponding to the allowed crystal reflections in the material, and which arise from the spatial distribution of atoms within a periodic lattice (Bragg scattering). As such, direct information on the character of the material can be obtained without further processing of the obtained diffraction pattern. In contrast, disordered systems display predominantly diffuse scattering – broad, ill-defined features in the F(*Q*) that correspond to the high degeneracy of local (and possibly long-range) ordering of objects in the system, and cannot generally be analysed without pre-processing.

A standard null-scattering $Ti_{0.68}Zr_{0.32}$ flat plate neutron cell with internal dimensions 35 x 40 x 2 mm with a wall thickness of 1 mm was used as the catalytic reactor. This particular alloy avoids adding significant structural contributions arising from the sample container to the measured data on atomic and molecular length scales, and contributes only a diffuse and modest Porod scattering component at low-Q arising from mesoscale compositional fluctuations in the alloy that can be readily accounted for when forming the total F(*Q*). The flat-plate geometry permits utilisation of NIMROD's maximum beam size at sample (30 x 30 mm) while limiting sample thickness in order to minimise multiple scattering and attenuation effects.