Designing Next Generation Emissions Abatement Catalysts Using Operando Neutron Total Scattering

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Integration of renewable feedstocks into the current energy infrastructure will require the development of catalysts and sorbents that can maintain high surface area and catalytic activity under challenging thermal/hydrothermal environments and acid gas (SOx, NOx and H2S) exposure. The design of new stable and acid-gas-resistant catalysts requires a deep understanding of sintering and acid gas interaction with active sites. Addressing these challenges will require advanced operando characterization of materials to effectively guide materials discovery. Studying the time-resolved structural evolution of materials under gas flow conditions is key to understanding catalytic performance under real-world operating conditions with the end goal of extracting design strategies for industrially relevant catalysts. Total scattering, including both Bragg and diffuse scattering signals, enables the study of structural evolution in catalysts and can provide key insights into how long-range, nanoscale, and local atomic structure motifs differ and deliver unique properties. Synchrotron X-ray scattering provides active metal site sensitivity and unprecedented temporal resolution, while neutron scattering offers light atom sensitivity and superior penetration of sample environments. We present in-situ studies following multiple length scales of interest in two very different catalytic material systems with these probes: (1) the oxidation and reduction behaviors of ceria nanorods at elevated temperatures, specifically following the nature of oxygen vacancies; and (2) sinter resistance, degradation, and regeneration behavior of novel high-entropy fluorite catalyst supports under acid gas exposure. Further, we will discuss the design and development of the hazardous gas handling system (HGHS) system under construction at the Nanoscale Ordered Materials Diffractometer (NOMAD) at the Spallation Neutron Source at Oak Ridge National Lab. The HGHS will deliver in-situ exposure to industrially-relevant acid gas at NOMAD, enabling investigations of acid gas interactions with sorbents and catalysts, which will be a unique capability among neutron sources in the world and will aid in the design of new materials and processes with higher energy efficiency and a smaller emissions footprint.