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X-ray absorption spectroscopy on catalysts, a photon tool to determine structure –performance relationsX-ray absorption spectroscopy on catalysts, a photon tool to determine structure –performance relations

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Catalysts are the enabling technology for conversion and synthesis of energy and chemicals. To increase the sustainability of chemicals and fuels production, new and better processes must be developed, which requires the development of new and better catalysts. Their design is greatly helped by the fundamental understanding of why one catalyst is a better one than another. To this end, in situ or operando (under working conditions) characterization is an indispensable tool. X-ray absorption and more recently, X-ray emission spectroscopy are versatile tools to determine the structure of a catalyst. Because of the large penetration depth of X-rays, notably in the hard X-ray regime, in situ and operando measurements are generally applied.
Heterogeneous catalysts often consist of nano-sized catalyst particles supported on a carrier. Often, dopants are added to increase activity, selectivity and / or stability. In a methane steam-reforming catalyst, rhodium supported on ceria-doped alumina, we elucidated the roles of the different catalyst components. The rhodium nano-particles are responsible for C-H activation of methane, which is the rate-limiting step; the role of dopant ceria is two-fold: the first is stabilization of the rhodium particles and the second water activation to generate oxygen that reacts to carbon atoms on the catalyst to prevent its further reaction to coke. In situ XAS measurements at the Rh K and Ce L3 edge combined with classical characterization tools and transient measurements enabled drawing a molecular level mechanism, which lead to the suggestion of improvement of the catalyst.

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