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Combining synchrotron characterization with transmission electron microscopy measurements for methane activation

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Methane is the main component of natural gas, being the simplest hydrocarbon that exists. Nevertheless, it is a very stable molecule (440 kJ·mol-1 are required to break a single C-H bond), therefore the development of highly active catalysts is required for methane conversion reactions. Different oxidizing agents for methane conversion can be chosen, such as oxygen for its oxidation to abate CH4 from tailpipes, or steam or CO2 in the steam reforming and dry reforming of methane, respectively, to generate hydrogen. The development of highly active catalysts requires the precise design of the catalytic active sites, which can only be achieved by characterizing the catalysts under their real working state. To that end, operando synchrotron measurements are required. Their combination with high resolution transmission electron microscopy measurements and, in particular, with in situ TEM studies can provide the full picture required to unravel how catalysts adapt and evolve under the reaction atmosphere (presence of gas and temperature).
In our projects related to methane abatement and methane dry reforming [1,2], we have combined in situ synchrotron X-ray diffraction (XRD), X-ray absorption spectroscopy (XAS), near-ambient pressure photoelectron spectroscopy (NAP-XPS) measurements performed at the ALBA synchrotron with ex situ high-resolution transmission electron microscopy (HRTEM) to study mono- and bimetallic nanoparticles (Pd, PdPt, ⋯) supported on ceria [3]. The synchrotron measurements allowed us to monitor the evolution of the bulk structure and the chemical environment of our catalysts'components as well as the surface oxidation state and its reorganization under reaction. The combination of these measurements with HRTEM observations allowed us to understand how the metals reorganize at the nanoscale and how the metal-support interface evolves during reaction. The synchrotron experiments revealed a gradual oxidation of Pd to PdO during methane oxidation, which paralleled an increase in methane conversion observed. By HRTEM, we observed a unique reorganization of Pd and Pt on the ceria surface after a reaction cycle at 1173 K under dry methane combustion conditions by forming Pt-Pd/PdO assemblies. These assemblies had a geometry similar that resembled that of mushrooms and we found that the mushrooms'feet were composed of PdO and the heads of PdPt. The HRTEM images provided very valuable information about the particular arrangement of Pd and Pt during reaction. A step forward in this study would be perfoming in situ TEM measurements. With these measurements, we could obtain direct observations on the different steps that lead to this unique reorganization of Pd and Pt on the ceria surface.

References

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