Identifying active sites in nanoshaped heterogeneous catalysts for energy and environmental applications.

Lluís Soler* and Jordi Llorca

Institute of Energy Technologies, Department of Chemical Engineering and Barcelona Research Center in Multiscale Science and Engineering, Universitat Politècnica de Catalunya, EEBE, Eduard Maristany 16, 08019 Barcelona, Spain

lluis.soler.turu@upc.edu

Heterogeneous catalysis is a very active discipline because it determines the success of many transformation processes. It is particularly relevant for both the heavy and fine chemical industries as well as in air pollution abatement generated by combustion engines and remediation of industrial exhaust gases, among others. Regarding metal/oxide catalysts, the emergence of different nanoshaped oxide supports should enable the study of the influence of the oxide morphology on the metal-oxide interaction and catalytic activity, but in spite of numerous studies in this field, the nature of the metal-support interaction remains a controversial issue. In parallel, operando methodologies have attracted a lot of attention over the last few decades, becoming a research field in the forefront of heterogeneous catalysis. The operando approach is an effective way to provide direct measurements of the influence of the reaction environment over catalytic materials, in order to identify the nature of the active sites, elucidate the mechanism of the reactions involved and decipher the relationship between catalytic performance and catalyst structure. Our recent findings illustrate how operando techniques such as near-ambient pressure X-ray photoelectron spectroscopy (AP-XPS) or X-ray absorption near edge structure (XANES) are excellent tools to monitor both the surface restructuring, chemical state and electronic structure of catalytic systems that critically depend on the reaction conditions. We conducted three main approaches to fabricate more efficient heterogeneous catalysts and photocatalysts with a focus on energy and environmental applications: (i) hydrothermal and solvothermal methods to synthesize nanoshaped oxides such as ceria or titania; (ii) 3D printing procedures to manufacture ceria or titania based monoliths and photocatalytic microreactors and (iii) mechanochemical methods to prepare heterogeneous catalysts with enhanced active sites. Our recent investigations showed the influence of the different nanoshaped CeO_2 supports on the oxidation states of the metals for the preferential oxidation of carbon monoxide (COPrOx) reaction [1] and their preferential segregation towards the surface during the steam reforming of ethanol (ESR) to produce renewable hydrogen [2]. We have also developed nanoshaped and 3D printed titania-based materials for the photocatalytic production of hydrogen under dynamic conditions using ethanol as electron donor [3–6], and our recent results of an unprecedented, nanometric amorphous shell around the ceria crystallites obtained by mechanochemical methods for soot [7] or methane oxidation [8].

References

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