Hard X-ray spectroscopy of molecular heterogeneous catalysts in mesoporous materials

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The goal of the CRC1333 is to perform heterogeneous catalysis in mesoporous materials by immobilizing molecular catalysts inside the pores and using confinement effects as key factor for selectivity. Both pore size and pore geometry are expected to have a significant impact on the reactivity as well as the coordination chemistry of the immobilized catalysts. In this context, spectroscopy with hard X-rays is used to analyze these influences based on the excitation of electrons close to the atom nucleus, mainly the 1s-electrons. Hard X-rays enable the investigation of the molecular nature of catalytic complexes fully independent of the state of aggregation. The portfolio of X-ray absorption spectroscopy (XAS) and X-ray emission spectroscopy (XES) consists of XANES (X-ray absorption near structure) to get information about the oxidation state of the central atom and EXAFS (Extended X-ray absorption fine structure) to illustrate bond distances as well as type and number of the coordinating atoms. Moreover, HERFD-XANES (high energy resolution fluorescence- detected XANES) and vtc-XES (valence-to-core X-ray emission spectroscopy) provide the HOMO- and LUMO energies of the complexes and ctc-XES (core-to-core X-ray emission spectroscopy) enables the identification of their spin states (Figure 1). The obtained data allow the comparison of the electronic and structural parameters of the molecular complexes in solution and inside the pores to get a deeper insight in the reaction mechanisms of the investigated catalytic reactions.[1-4]

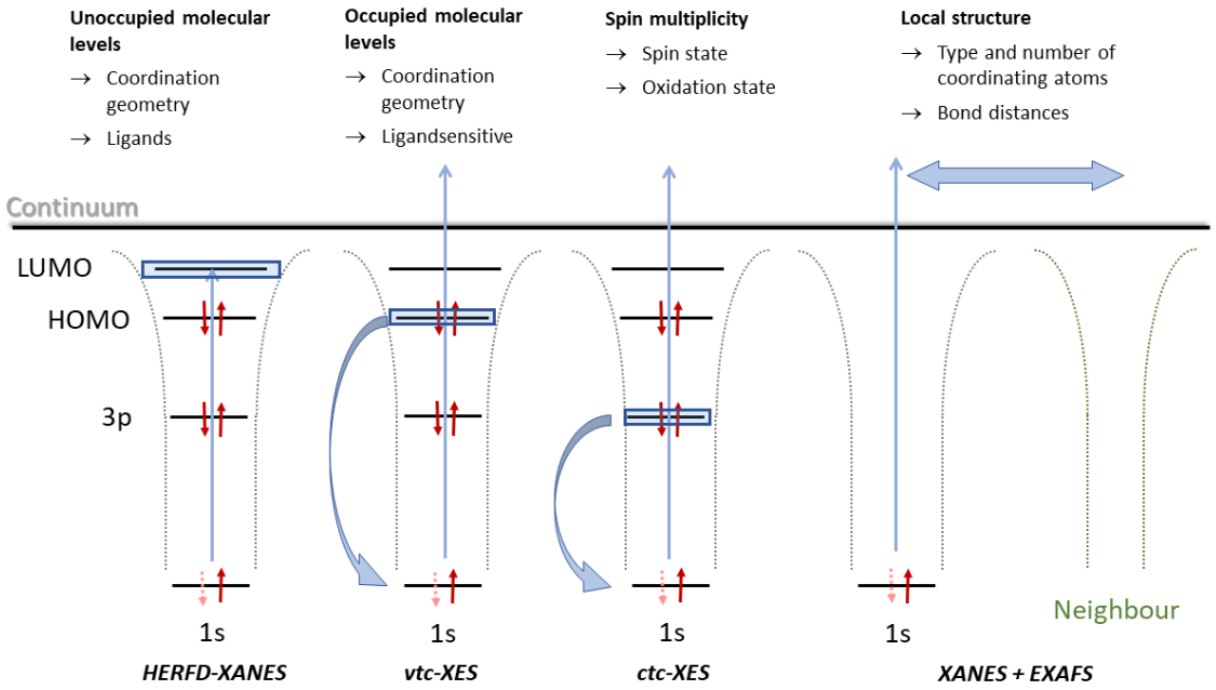

Figure 1: Hard X-ray methods and provided information for the example of K-edge spectroscopy.[5]

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