

Dynamics of a Pt/Al₂O₃ catalyst in the presence of H₂: complementary results from INS, FT-IR and DFT

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Supported platinum nanoparticles are largely employed in industrial hydrogenation processes thanks to their great catalytic activity which, in turn, derives from their ability to activate and split the H₂ molecules at the metal surface to form reactive hydride species. Albeit these systems were among the very first heterogeneous catalyst to be discovered and employed, many details about their behaviour under reaction conditions are still matter of debate and subject of extensive research effort.

The sample we have been focusing on is characterized by a very large dispersion of the Pt nanoparticles (1.4 ± 0.4 nm), which are supported on a high surface area transition alumina. The sample was prepared in the laboratories of the industrial company Chimet S.p.A. and subjected to an extensive characterization work, aimed to better understand its behaviour under hydrogenation conditions.

Among the employed techniques, Inelastic Neutron Scattering (INS) spectroscopy had been fundamental, thanks to its great sensitivity towards the vibrational modes of all H-containing species. The INS spectra of the sample were measured under a high and low H₂-pressure on the IN1-Lagrange instrument at the ILL, resulting in significantly different spectral profiles [1]. The spectra were interpreted on the basis of an extensive DFT simulation work comprehensive of 26 different Pt_xHy/γAl₂O₃ models, performed in collaboration with IFPEN. The comparison clearly indicated that under high H₂-pressure conditions the Pt nanoparticles mostly reconstructed into cuboctahedric morphologies in weak interaction with the support, while the spectral profile collected at low H₂-pressure is compatible with less symmetric morphologies where the nanoparticle is in stronger interaction with the support. In the latter case, the spillover of some H onto the support and the following formation of surface -OH groups was also observed. Complementary information was obtained by means of FT-IR experiment performed in operando conditions, allowing to follow the evolution of the system upon the progressive decrease of the H₂ concentration. In this latter case, the spectral evolution clearly indicated the conversion of multi-folded Pt-H species into linear ones upon their removal [2]. Both these findings are in good agreement with previous AIMD simulations, predicting the reconstruction of Pt₁₃ nanoparticles supported on γ-Al₂O₃ as a function of the temperature and the H₂ equilibrium pressure [3].

Overall, this study provided important new complementary information on the dynamic behaviour of Pt/Al₂O₃ catalyst under hydrogenation reaction conditions, representing a new important step in the understanding of their real nature.

References

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Primary authors: VOTTERO, Eleonora (Università di Torino); Dr CAROSSO, Michele (Università di Torino); Mr RICCHEBUONO, Alberto (Università di Torino); JIMENEZ-RUIZ, Monica (ILL); Dr PELLEGRINI, Riccardo (Chimet S.p.A.); Dr CHIZALLET, Céline (IFPEN); Dr RAYBAUD, Pascal (IFPEN); Prof. GROPPPO, Elena (Università di Torino); PIOVANO, andrea (ILL)

Presenter: VOTTERO, Eleonora (Università di Torino)

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