PhD thesis proposal for funding by the Doctoral School of Chemistry
University Claude Bernard Lyon 1-Year 2019

Structural dynamics of Pt/CeO_2 catalysts for hydrogen production

In the near future, dihydrogen could be widely used as a carrier of renewable and clean energy for transport and electricity generation. H_2 is currently produced in syngas form ($CO + H_2$) from fossil fuels, biomass or wastes as raw ressources [1-3]. The water-gas shift reaction ($CO + H_2O \rightarrow CO_2 + H_2$) allows reducing the CO content in H_2. Generally, Fe-Cr and Cu-Zn catalysts are used for the reaction carried out at high and low temperature, respectively. In order to achieve higher conversion rates, recent research has focused on CeO_2-supported metal catalysts (Pt, Pd, Cu) because of the unique redox properties of this oxide [4].

The design of new catalysts needs to take into account the scarcity of elements, especially noble metals. When reducing the metal content in a catalyst, particular catalytic species (isolated atoms, subnanometric clusters) can be stabilized [5]. We have recently shown that the structure and performances of Pt/CeO_2 catalysts can be strongly improved by specific conditioning based on alternating oxidizing and reducing atmospheres at moderate temperatures [6]. A precise tuning of the structural dynamics of the catalysts (Figure 1) favors the formation and stabilization of more active metal nanoparticles for catalysis.

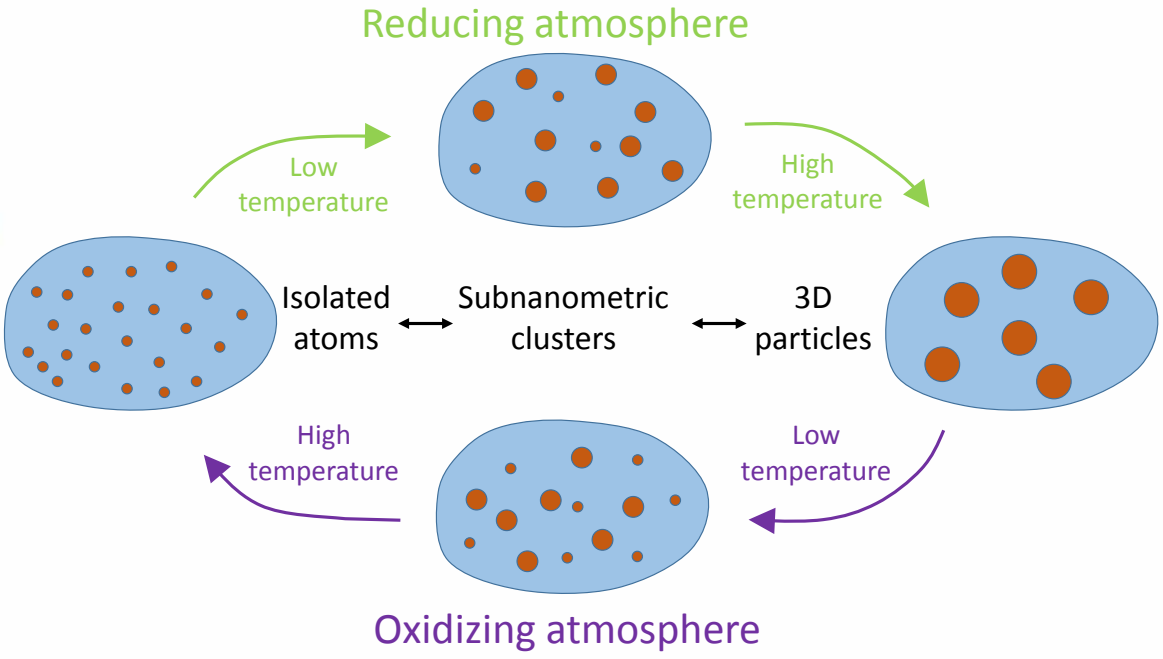

Figure 1. Structural dynamics of a catalyst consisting of metal particles (orange) supported on a reducible oxide (blue).

IRCELYON • UMR5256
2 avenue Albert Einstein • F69626 Villeurbanne Cedex
tel 33[0] 472 445 300 • fax 33[0] 472 445 399
http://ircelyon.univ-lyon1.fr

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The main objective of this PhD thesis work is to unravel the mechanisms of structural dynamics of Pt/CeO_2 catalysts, with application to water-gas shift for energy production. The scientific approach is based on a coupling between catalytic/kinetic measurements and advanced *in situ/operando* characterization techniques (electron microscopies, Raman and fluorescence imaging, infrared and X-ray absorption spectroscopies). In particular, attention will be paid to the location (surface, sub-surface, grain boundaries) and scale (nanometric, micrometric) of the involved phenomena and the specific role of H_2O.

The thesis will be carried out at the Institut de Recherches sur la Catalyse et l'Environnement de Lyon (IRCELYON) in the ECI2D team. The M2 student must have a strong scholar background including heterogeneous catalysis, characterization techniques and physical chemistry of materials. In addition, high scores in L3, M1 and M2 exams (or equivalent) are required by the Doctoral School of Chemistry of the University Claude Bernard to obtain a scholarship. Motivation and curiosity for scientific research associated with a critical mind is required.

Contacts:

- PhD thesis director: Dr. Stéphane Loridant (IRCELYON), Tel: 0472445334, E-mail: stephane.loridant@ircelyon.univ-lyon1.fr

- Co-director: Dr. Laurent Piccolo (IRCELYON)

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