PhD project

In-situ and operando Transmission Electron Microscopy of catalysis reactions for methane conversion

Keywords: in-situ Transmission Electron Microscopy, Catalysis, Zeolites, Methane conversion

Expected starting date: as soon as possible, in any case before end of 2020

Location :
Groupe de Physique des Matériaux
University Rouen Normandy
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Supervisors: X. Sauvage and S. Moldovan
Partner: LCS Caen (https://www.lcs.ensicaen.fr/en/)

Requirements:

The candidate should have a master in physics, chemistry or material science. A background in materials physico-chemical characterisation methodologies adapted to nanomaterials would be an asset, especially in electron and/or optical microscopy. Theoretical and practical knowledge on the electron-matter interactions and detection methods will be also a strong advantage.

Description:

In the actual context of the significant demographic evolution as we are assisting at the diminution of the natural resources, the demand of solutions for ultra-clean alternative fuels and chemicals is rapidly growing. The methane conversion offers an attractive solution due to the availability of cheap natural gas with relatively low carbon footprint, as a cleaner alternative of the fossil oil and coal largely used today. Situated at the core of energy related chemical reactions and processes, the catalysis relies on complex methodologies and applications. Since in heterogeneous catalysis, complex materials with properties defined at the nanoscale are commonly employed, the complete understanding of the catalysts behavior in the course of reaction by Transmission Electron Microscopy (TEM) combining the *in-situ* and *operando* methodologies is essential to bridge the materials processing with their performances.

The main objective of this PhD project relies on environmental TEM that allows to probe the microstructural and changes within zeolite-based catalysts in the range of temperatures up to 1000°C and at atmospheric pressures under well controlled gaseous environments. This experimental setup will be employed to explore the evolution and performances of zeolite crystals after the incorporation of metallic heteroatoms (Cu, Fe, Ti, Mo, W, …) prone to highly increase the activity for CH_4 conversion. The 2D and 3D TEM investigation of zeolite-based materials is rather challenging due to the high sensitivity of this class of materials to the electron beam. On the one hand, the electron tomography essays (3D imaging) are based on the reconstruction of volumes using stack of images, which imply quite long durations of specimen exposure to the electrons. The use of high acceleration voltages triggers the ionization of the gas in the cell, which would certainly modify the materials behaviour under reaction conditions, on the other hand. In this context, the first step of the PhD project will be dedicated to the definition of the experimental conditions to be employed for the 2D and 3D TEM investigations. Special care will be given on the investigation of the influence of the electron dose and the acceleration voltage to avoid or reduce sample damages. The double corrected TEM ARM200-CF of Genesis platform will be used for this study, which brings the advantage of reducing the acceleration voltage while keeping the atomic resolution of the TEM. Concerning the 3D characterization of zeolite materials, the emphasis will be put on the setup of fast tomography approach by using a fast camera and the adaptation of the compressive sensing methodology for 2D and 3D imaging. The 2D microstructural and chemical changes of zeolite crystals under CH_4 environment and conditions required for methane conversion will be explored, with particular focus on the impact of the nature and distribution of the metallic heteroatoms in the zeolites. Such experiments will allow the choice of a system suitable for deeper investigations. The first system to be explored is the Mo impregnated zeolite in order to determine the correlation between the microstructural changes and its catalytic activity. The impact of electron beam on the microstructure changes will be investigated in parallel by taking advantage of the multiple transparent areas within the E-cell.

Since the active surface and the accessibility to the active phase plays a key role for the catalytic performance, these features will be studied for a heterogeneous catalysts with high catalytic activity, electron tomography experiments will be carried out on the very same or similar specimen at different stages of the reaction . One of the original parts of the current project is the investigation of new large pores zeolites synthesized by our partner LCS. conversion is carried out.

Application:

Candidates should send a CV, a motivation letter and recommendations (if any) to:

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