Rietveld refinement of XRD-CT data

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The main aim of solid catalyst study is to understand the relation between catalyst structure and its function. The analysis of solid catalyst structure under operando conditions can provide the user with essential information about the catalyst active sites[1].

X-ray Diffraction Computed Tomography (XRD-CT) allows for imaging the interior of a catalyst, simultaneously providing the information about its chemical composition. When combined with a bright X-ray beam and state-of-the-art detectors with a fast data acquisition/data readout, this technique can be applied to study the three-dimensional structure of catalysts, under operating conditions[2].

When analysing a diffraction pattern of a crystalline material containing several phases, one may find difficulties in the phase identification, due to the peak overlap. Therefore, the Rietveld refinement should be performed, aiming to refine the model structure of the catalyst, created by the user. In addition, for each phase presented in the model, useful information regarding the lattice parameter, crystallite size or finally percentage composition can be extracted. However, analysing diffraction patterns as a function of an external variable, such as temperature or pressure, leads to better understanding of the behaviour of the system under investigation. This approach, called parametric refinement, in comparison with single or sequential refinement, results in higher precision of refined parameters and greater stability of refinement process[3]. In addition, it offers a possibility to refine the non-crystallographic factors, such as temperature, which can be useful when treating the space resolved data, where the possibility of hot-spots formation is possible.

In this study, a La-Sr/CaO catalyst, used in OCM process (Oxidative Coupling of Methane) was studied in a packed-bed reactor. The catalyst was firstly placed in the atmosphere of neutral gas during the temperature ramp (from room temperature to 780 °C) and then in the mixture of methane and oxygen, with different methane to oxygen ratio. The XRD-CT analyses were performed at the station ID31 of ESRF in Grenoble, using a monochromatic X-ray beam of 70keV. The data was refined using software Topas 5 and MATLAB.

This study aims to present the approach of parametric refinement of XRD-CT data and to demonstrate the applied methodology using the obtained preliminary results to explain it.

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