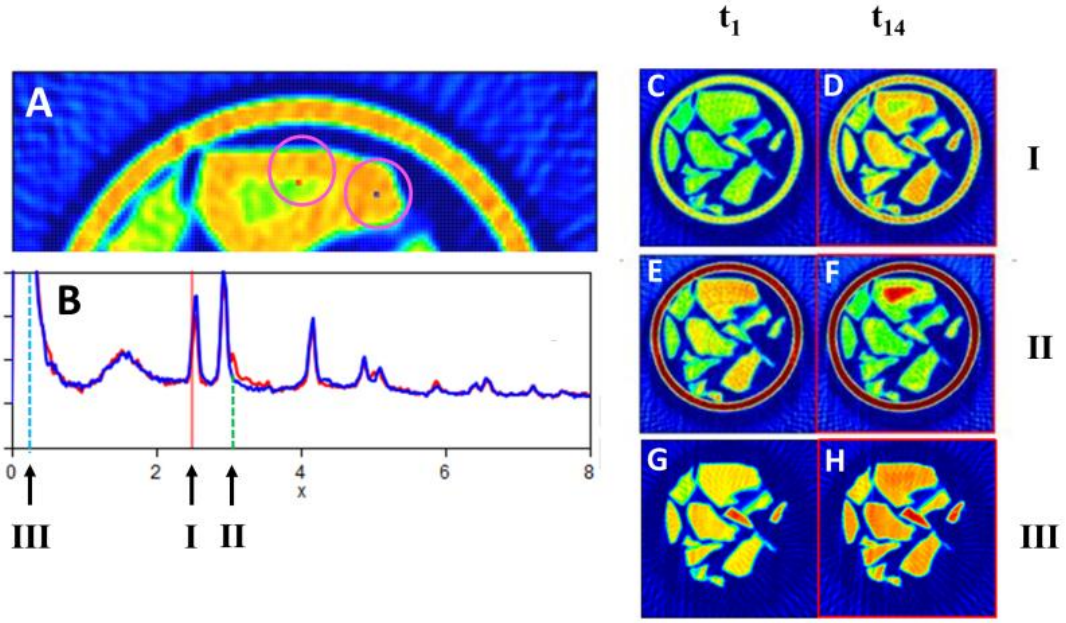
CH3726: Development of PDF tomography
S.D.M. Jacques et al.

The experiment CH3726 was concerned with the development of Pair Distribution Function Computed Tomography (PDF-CT); our initial development work (a study of Pd nanoparticles on alumina) has been submitted and provisionally accepted for publication[1]. Below we report results from the initial analysis of data collected during the *in situ* study of an industrial Co catalyst. This catalyst is used used in the Fischer-Tropsch (FT) synthesis of long chain hydrocarbons from synthesis gas (a mix of CO & H_2). We followed the calcination and activation of the catalyst and attempted an *in operando* FT synthesis, albeit at low gas pressure for short chain alkane production, using PDF-CT.

The Co catalyst under study is supported on silica, manufactured as beads of non-uniform shape and size (typically 10-50 μm). These were packed into a 400 μm capillary (to mimic an industrial packed bed) and mounted to a gas delivery study. The capillary was heated using gas blowers. Temperatures and gas flow rates were set to replicate the industrial processing and operating conditions. PDF-CT was used to monitor the chemistry at one fixed slice in the bed. The figure below shows some reconstructed diffraction data collected at this slice during the course of the *in operando* study. The data requires further treatment to arrive at the PDF transform but these initial results show that catalyst activity is severely impaired by insufficient formation of metallic Co during activation and that this deactivation has a spatial dependence. However, there are other proposed deactivation routes (such as formation of carbide phases under operating conditions) and we hope that the PDF will resolve the pathways of these if they are shown to occur and whether they are surface or bulk



In operando study of industrial FT catalyst using XRD/PDF-CT. The latter was used to monitor one slice of the bed over time to see the changes in chemistry and physical state during the first few hours of operation of the catalyst. Maps such as that shown in A have been simply made from raw intensity components of the reconstructed diffraction for this slice. B shows two reconstructed patterns blue and red spectra from the pixel positions circled in A. The maps C-H are maps of raw diffraction intensity at positions I, II and III in the diffraction pattern; where I and II correspond to positions associated with Bragg peaks for CoO and fcc Co respectively. Position III corresponds to a SAX signal which can perhaps be rationalised with the PDF transform later. C,E & G and D,F & H show these components at times t_1 and t_{14} respectively. These images illustrate that spatial changes in time can readily be observed. The reconstructed diffraction patterns can be analysed using standard methods to enable mapping of phases and other information such as crystallite size.

[1]*Pair Distribution Function Computed Tomography.* S. D. M. Jacques , M. Di Michiel, S. A. J. Kimber, X. Yang, R. J. Cernik, S. J. L. Billinge and A. M. Beale. Accepted subject to editorial changes. Nature Communications (2013).