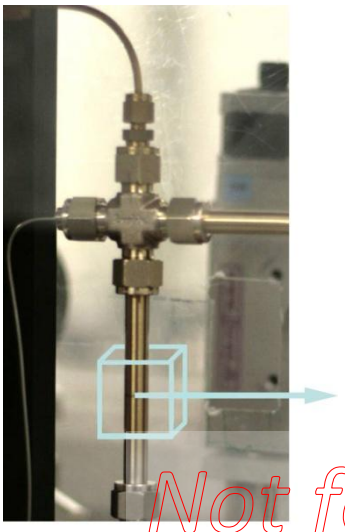
Imaging a supercritical water crystallisation vessel using in-situ diffraction

Our aim in this experiment was to use a combination of high-energy angle- and energy-dispersive diffraction to probe the interior of a continuous hydrothermal flow synthesis (CHFS) reactor *in-operando*. CHFS provides a route for the controlled synthesis of nano-particle materials. The work is a continuation of work carried out in MA387, here the aim was to investigate the synthesis of hydroxyapatite (HA) and ceria under a number of conditions. The beam-time took place June/July 2008 so the data is still being processed. Nevertheless, a few preliminary results are shown below.

The setup was that similar to that we achieved in MA-387. The CHFS reactor was mounted on a translational stage in order to carry out spatial angle-dispersive and energy-dispersive mode diffraction (which requires with switching between monochromatic and white beams). Figure 1 shows some selected results from HA synthesis using diammonium hydrogen phosphate and calcium nitrate tetrahydrate. We were worried that the signal from this phase was low, so only carried out a limited number of studies with this material and then switched to the synthesis of ceria from cerium ammonia nitrate. Figure 2 shows the intensity maps, i.e. the distribution of the phase, as determined by angle-dispersive diffraction at three synthesis temperatures which clearly show differences.



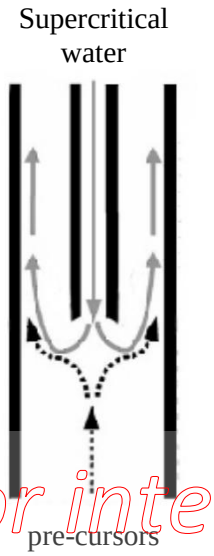
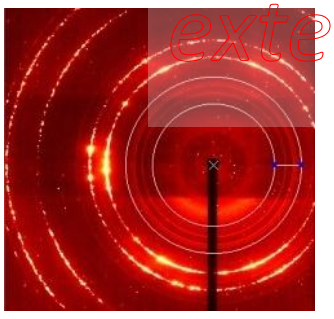
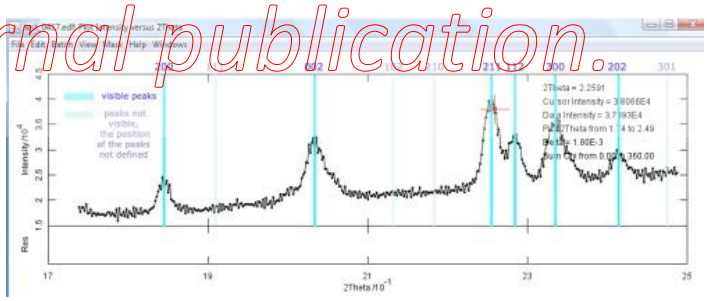
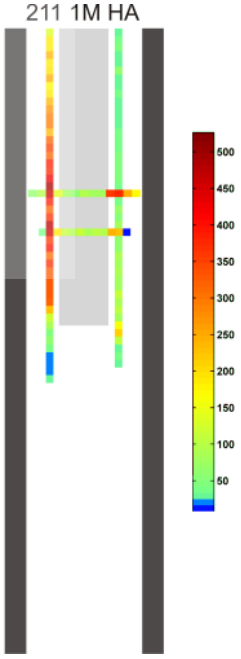


Figure 1 Photograph (A) and schematic (B) of CHFS reactor. The reactor is spatially scanned and at each scanned point, diffraction is collected in the chosen mode; here, single 2D powder ADD pattern (C). This is integrated to obtain a conventional 1D powder (*D*); overlaid are the reflections positions for HA. Intensity and peak widths can be mapped. (E) shows the 211 HA reflection over 4 traverse lines (two vertical and two horizontal) inside the steel CHFS reactor; here the steel walls are indicated.







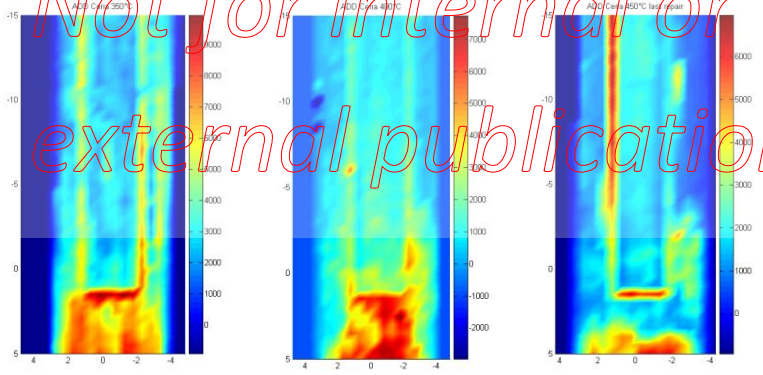


Figure 2 Ceria distribution in its synthesis at three temperatures: 350°C (sub-critical), 400°C (critical), 450°C (super-critical) shown from left to right respectively. These images have been derived from angle-dispersive data

At super-critical temperatures the majority of material forms away from the inner nozzle. We will construct similar maps of crystallite size (determined from peak width analysis). These results and those derived from experiment MA387 (which are under review for publication in a leading journal) represent the first look inside a functioning CHFS reactor. This information is critical to the understanding and design of both CHFS processes and reactors.

Dr Simon Jacques. September 2008