	Experiment title: Development of molecular structure and organisation during the mercerisation of cellulose	**Experiment number**: SC-1389
Beamline: ID02A	**Date of experiment**: From: 9/7/2004 to: 12/7/2004	**Date of report**: 27/7/2004
Shifts: 9	**Local contact(s)**: Dr. T Narayanan	*Received at ESRF:*

Names and affiliations of applicants (* indicates experimentalists):

A Mahendrasingam*, AK Wright, M Parton*, J Rasburn*, DJ Blundell and W Fuller

School of Chemistry and Physics, Keele University, Staffs, ST5 5BG, UK

UCB Films, Wigton, Cumbria CA7 9BG, UK

Report:

The purpose of this investigation was to utilize the high-brilliance of the ID02 beam-line and in advanced time-resolved methods to follow the molecular reconfiguration during the mercerisation of native cellulose.

The experiments preformed used native cellulose pulp supplied by Surface Specialties and also Whatman cellulose I filter paper. The time-resolved SAXS/WAXS data were recorded during the in-situ mercerisation on beam line ID02 using the ESRF CCD detectors with a typical exposure time of 0.05 seconds per frame and a gap of 0.95 seconds to download the data. The mercerisation was fully completed within 100 frames. A number of mercerisation experiments were preformed using a variety of NaOH concentrations and at various temperatures.

Figure 1 shows a time vs. two-theta scans for a mercerisation experiment at room temperature using a NaOH concentration of 25%. The time scale for the experiment is 100 seconds (the top represents the first X-ray exposure and the bottom represents the final X-ray exposure). It can be seen that cellulose I initially transforms in to a poorly ordered crystalline Na-cellulose II

complex as shown by point A in figure 1. Cellulose II structure is regenerated from this complex during the subsequent treatment of the sample with water.

The crystallinty of the regenerated cellulose II depends on post treatment of the mercerised Na-cellulose complex. By carefully controlling the mercerisation and post treatment of the Na-cellulose complex it should be possible in future experiments to control the level of crystallinty and the polymorph. These observations are supported by recent work by Nishiyama et al, J,Wood.Sci (2000). Sarko et al (1985) reported the presence of numerous intermediate polymorphs of during the mercerisation process. In our present study we have shown that the occurrence of a particular type of polymorph depends on the temperature and concentration of the NaOH. For example cellulose IV polymorph was only present at a specific range of NaOH concentrations and temperatures.

The preliminary analysis of the time-resolved SAXS data shows a definite structural change in the long range order during the mercerisation. Preliminary SAXS analysis provides information on the size and degree of anisotropy of pores in the film; it appears that there is a reorganization of pores and their size during the mercerisation. Nishiyama et al (2000) showed that at a certain temperatures during mercerisation the Na-Cellulose II complex can revert back to cellulose I rather than adoption of the cellulose II configuration.

These results clearly demonstrate that we could control the final level of crystallinty and the type of polymorph by carefully controlling the concentration and temperature of the mercerisation and post processing of the cellulose. Therefore we are proposing to continue this work by using the flow cell, which is available on ID02 to control the concentration and temperature on NaOH during the mercerisation and post processing.

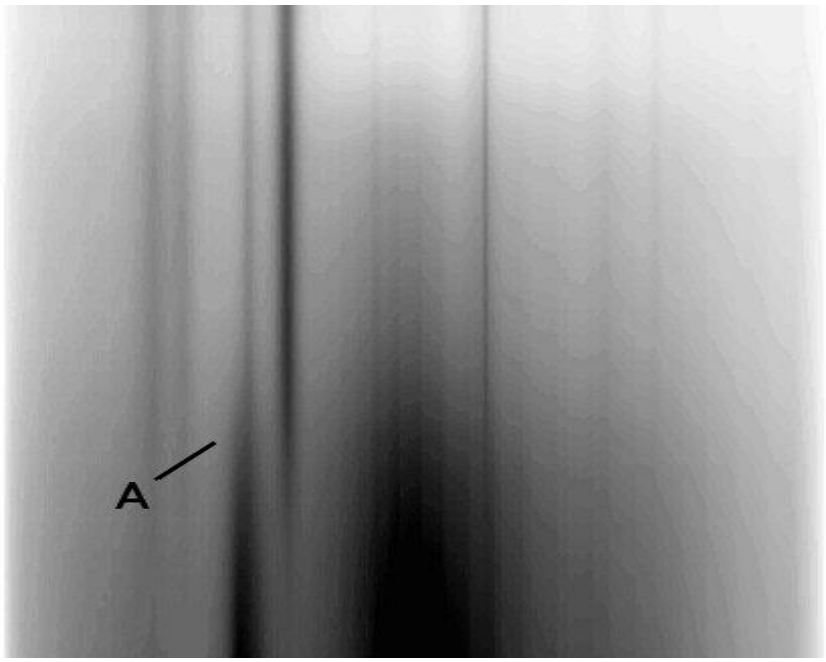

Figure 1: Time versus two-theta scan for a mercerisation
Experiment at room temperature using 25% NaOH

[1]A.Sarko et al, Crystalline Alkali-Cellulose Complexes as Intermediates During Mecerisation, Ch 9 (1985); [2]Y.Nishiyama, S.Kuga, T.Okano, J,Wood.Sci (2000) 46:452-457