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Tensometry of carbon fibres and elastomers at the diffractometer at BL20B of the Photon Factory

D. C. Creagh,[a]* P. M. O'Neill,[b] R. J. Mills[b] and S. A. Holt[b]

[a]*Information Sciences and Engineering, University of Canberra, Belconnen ACT 2616, Australia, and* [b]*School of Physics, University College UNSW, Canberra ACT 2600, Australia. E-mail: d-creagh@adfa.edu.au*

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Two systems have been developed for the simultaneous recording of the SAXS and the WAXS patterns from carbon fibre and elastomer samples which are placed under stress. The systems have been designed to fit inside the versatile vacuum diffractometer (BIGDIFF) at the Photon Factory. In one system, use is made of the ability to move the imaging-plate cassette. In the other, use has been made of an imaging-plate changer which can deliver up to 13 plates into position with a duty cycle of about 60 s. In this case each imaging plate can record SAXS/WAXS patterns in the range 0.5–20° due to the passage of the beam through the specimen which is mounted in a specially designed tensometer. Because BIGDIFF is a vacuum diffractometer and parasitic scattering is small, exposure times as short as 2 s can give acceptable SAXS/WAXS patterns. The systems have been used for the study of both the change of structure with strain, and the relaxation processes which occur as a result of the sample being strained at a fixed rate by a predetermined amount.

Keywords: X-ray diffraction; carbon fibres; elastomers; tensometry; SAXS; WAXS.

1. Introduction

The Australian National Beamline BL20B at the Photon Factory comprises a primary slit system, a primary monochromator system, white-beam beamstop and beam shutter (Cookson *et al.*, 1992). Two monochromator configurations are available: a tunable Si(111) channel-cut system and a fixed-exit-height double-crystal system with sagittal focusing. Sagittal bending of the second monochromator crystal can give an increase in photon flux of up to 25 times (Creagh & Garrett, 1995).

Within the experimental hutch is the versatile vacuum powder diffractometer BIGDIFF (Barnea *et al.*, 1992). BIGDIFF is primarily a powder diffractometer, with imaging plates as the preferred means of recording diffraction patterns. Within it a wide variety of devices can be mounted. One of these, a tensometer for the correlation of material structure with strain, is the subject of this paper. Experiments have been performed with or without the Weissenberg slit installed in BIGDIFF (Mills, 1995) for SAXS experiments or with a multiple-plate imaging-plate changer installed (Creagh *et al.*, 1997) for SAXS/WAXS experiments.

Gas-filled two-dimensional detectors have been used in some experiments we have undertaken in other laboratories. These we have found to be significantly slower than imaging plates for SAXS/WAXS experiments, and the fast relaxation effects we wish to observe are long finished during the course of an exposure (Creagh *et al.*, 1997; O'Neill, 1997). We plan SAXS experiments at SRI-CAT at the Advanced Photon Source using fast CCD detectors.

2. Equipment

2.1. *Small-angle X-ray scattering configuration*

BIGDIFF was built with the facility for translation of the imaging-plate cassette as an integral part of the design. The entire cassette can be translated across the incident beam by the width of an imaging plate under computer control using the same *SPEC* software as used by all the motor encoders in the BL20B system. Also, a pair of Weissenberg screens can be fitted to BIGDIFF to expose only a relatively small strip (10 mm wide) of the imaging plates (Creagh *et al.*, 1998). The diffractometer can be used with or without the Weissenberg screens in position. For the SAXS experiments the monochromator was used in the unfocused mode, with the beam dimensions limited by secondary slits upstream of the tensometer. The tensometer was mounted on the θ axis of the Huber 410/420 two-circle diffractometer system. For long-time exposures, the Weissenberg slit system was used to protect the imaging plate from scattered radiation. For experiments of less than 1 h total duration, a simpler arrangement was used consisting of a rectangular lead mask, 210 × 135 mm, in which a rectangular aperture of dimensions 160 × 110 mm was cut. This was mounted on a frame located on the 2θ arm of the diffractometer. The mask served two purposes: to reduce scattered radiation and to act as the support for the lead beamstop (8 × 8 × 8 mm), which was mounted at the centre of the aperture.

Samples of the fibres under test were mounted on specially made test cards (ASTM, 1989). For the purpose of alignment, burn paper was fixed to one of the test cards behind one of the fibres and burns were taken as the upstream slit was translated across the sample, using a slit width of 150 μm. When the burn mark was symmetrical with respect to the sample, a pinhole was made in the paper and the fibre was cut away. The alignment laser for BIGDIFF was then set up such that the beam passed through the pinhole and passed also through the upstream slit. The jaws of the upstream slit were then closed down symmetrically about the beam to a width of about 50 μm. Then the 2θ arm carrying the beamstop was rotated so as to cut off the laser beam. A long exposure burn was then taken in front of and behind the beamstop to confirm that the X-ray beam was in the correct position.

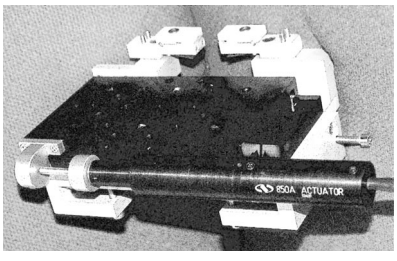


Figure 1
Photograph of the tensometer. The Newport 50 mm travel linear slide is driven by a Newport 850A actuator. At the top of the photograph the jaws of the tensile stage can be seen. These conform to ASTM Standard D3379.

Careful positioning of the beam is essential since the fibres which have been tested have diameters of 25 μm or less.

The tensometer is mounted on the flat-plate spinner mount of BIGDIFF. It has screws for the adjustment of the fibre in the vertical direction with respect to the beam. Fig. 1 is a photograph of the tensometer. It is based on a Newport 50 mm linear slide driven by a Newport 850A linear motor encoder. The position of the actuator is controlled using standard instructions from the *SPEC* operating system of BIGDIFF. Positioning is possible to a precision of 1 μm.

The SAXS pattern is recorded on an imaging plate mounted in the normal imaging-plate position (573 mm from the axis of rotation of the diffractometer). The beam shutter is used to shut off the beam during translation of the imaging-plate cassette. Five SAXS patterns were recorded on an imaging plate during a tensile test run.

2.2. *SAXS/WAXS configuration*

Fig. 2 is a photograph of the SAXS/WAXS configuration for the tensometer. For this configuration the sagittal focusing monochromator focused the synchrotron radiation at the plane of the imaging plates. The conventional imaging-plate operation was replaced by a multiple-plate imaging-plate changer (Foran *et al.*, 1998), and the beamstop with a smaller device mounted directly on the Huber top plate. The distance from the tensometer to the imaging plate is 303 mm. The imaging plates are set so as to record the top left-hand quadrant of the SAXS/WAXS pattern. Under these conditions, WAXS patterns up to 30° can be observed. The smallest angles observable are determined by the size of the beamstop. Typically, the SAXS patterns of polyurethane samples can just be resolved ($Q = 0.02$ A^{-1}; $2\theta = 1.5°$). In our initial experiments, exposure times for polyurethane linear segmented co-polymers with a (hard–soft)^n-type structure (Meijs *et al.*, 1996) were 30 s per increment of strain, and strain rates of between 66 and 366% per minute were used.

For later experiments, studies of the relaxation processes which take place after rapid straining were undertaken. For these the exposure time was reduced to 2 s with no apparent degradation in image quality (Yozghatlian *et al.*, 1997).

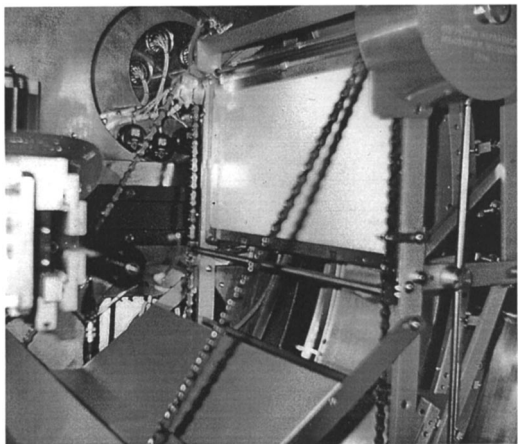


Figure 2
Photograph of the tensometer in the SAXS/WAXS configuration. The tensometer is on the left of the photograph, and the imaging-plate changer can be seen on the right. The imaging plate is set to give the upper right-hand quadrant of the SAXS/WAXS pattern.

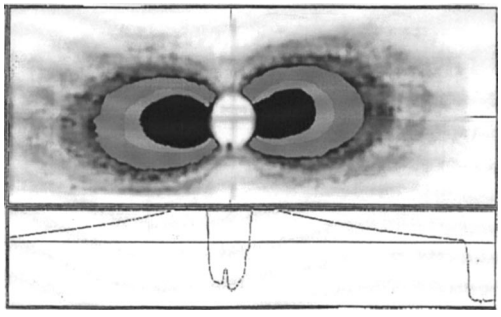


Figure 3
Small-angle scattering pattern of a carbon fibre 25 μm in diameter at 1% strain. Intensity scans through the symmetry axes are shown on the left of, and below, the SAXS pattern.

3. Results and conclusions

SAXS studies have been made of carbon fibres (polyacrilonitride heated at 300 K) and a nylon-related polymer.

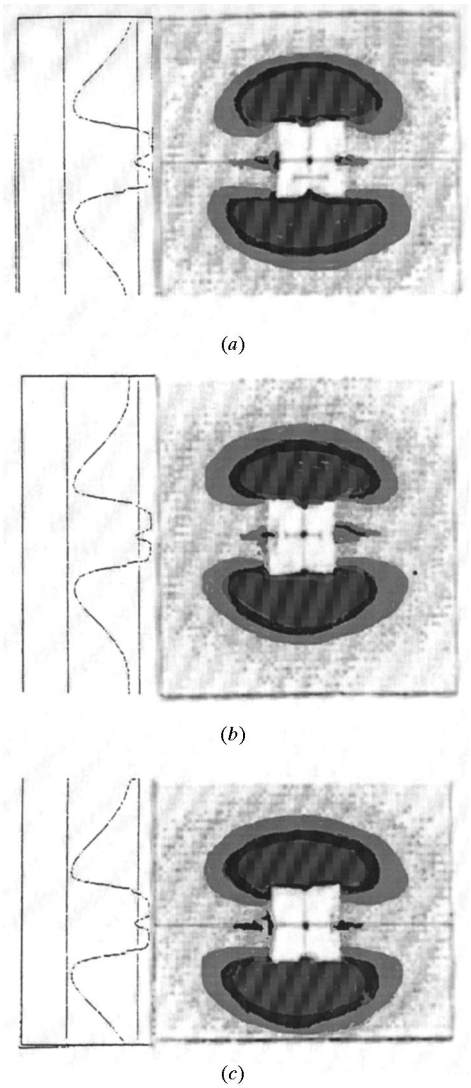


Figure 4
Small-angle scattering patterns from nylon-related thread 25 μm in diameter at (*a*) 6% strain, (*b*) 12% strain and (*c*) 18% strain. Intensity scans through the symmetry axis are shown on the left of each SAXS pattern.

For the carbon fibre the maximum strain which could be imposed before failure was 1%, consistent with the known stiffness of this material (Young's modulus = 120 GPa). This corresponded to a 1% change in the measured Q ($= 4\pi \sin\theta/\lambda$) value for the SAXS peak. The SAXS pattern for the carbon fibre near 1% strain is shown in Fig. 3.

For the nylon-related polymer the rate of change of the Q value of the SAXS peak was -0.016 Å^{-1} per 1% strain for strains up to 3%. In this region the material was essentially elastic. Measurements were taken to a maximum strain of 22%. The rate of change of Q with nominal strain was -1.07 Å^{-1} per 1% strain for this region. No yield drop was observed for this material. Fig. 4 shows a sequence of three SAXS patterns (6, 12 and 18% nominal strains) of the 25 μm-diameter fibre in the second stage of deformation.

For the linear segmented polyurethanes the SAXS/WAXS results of a cycle of extreme tensile testing are shown in Fig. 5. For this material (Creagh *et al.*, 1997) the specimen was subjected to up to 320% strain and then the stress was slowly relaxed to zero.

(i) At zero strain this material is highly crystalline (Fig. 5*a*) and an approximately Hookian stress/strain relation exists below 20% strain ($E = 2.5 \times 10^7$ MPa).

(ii) For the subsequent 130% strain the modulus dropped to about 4.0×10^6 MPa. During this phase the material loses crystallinity, although some crystallinity may be seen at 80% strain (Fig. 5*b*).

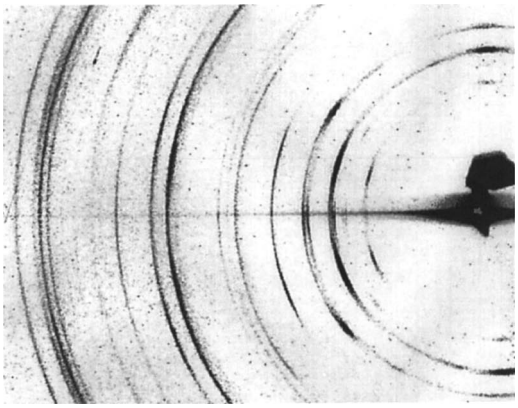
(iii) Thereafter the modulus increased to 1.0×10^6 MPa and the material becomes completely amorphous (Fig. 5*c*).

(iv) On releasing the stress, the stress/strain relation is different to that when the stress is initially applied, and at zero stress a permanent set of 100% is observed. The material has reverted to its original crystalline state. It does not remain in this state but slowly reverts to its original dimensions over the course of a month.

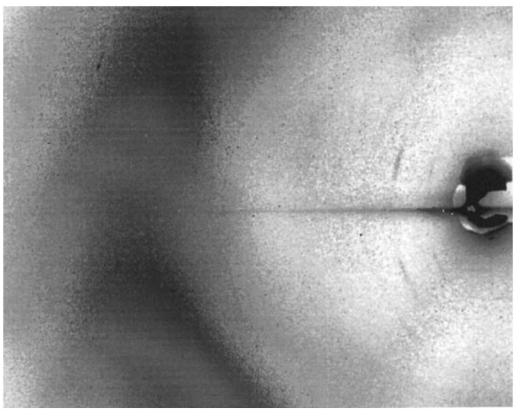
Three different relaxation processes have been identified, and a study of how these relate to changes in morphology is in progress. Preliminary investigation (Yozghatlian *et al.*, 1997) shows that a rapid (<1 s) relaxation is followed by a slower relaxation (~1 h), and a subsequent reversion to the original state over a period of a month.

In summary, we have developed a tensometer for use at the Australian National Beamline (BL20B) at the Photon Factory which is capable of studying the relation between structure and strain in a range of polymeric materials. A number of improvements are being made to the system, including the incorporation of a load cell, the construction of beam stops which are smaller and which can be positioned more precisely, and the installation of a more powerful linear actuator to allow faster rates of strain.

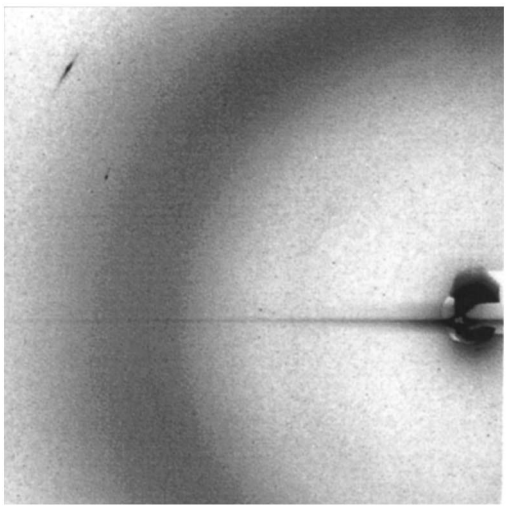
The system is small, and it will be possible to continue the study of the fast relaxation processes using fast two-dimensional detector systems at the Advanced Photon Source, where the Australian Synchrotron Research Program is a member of two consortia, CHEMMATCARS and SRI-CAT.



(*a*)



(*b*)



(*c*)

Figure 5
SAXS/WAXS patterns for a linear segmented polyurethane specimen under extreme tensile testing: (*a*) initial state, zero strain; (*b*) 80% strain; (*c*) 320% strain.

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