JOURNAL OF MATERIALS SCIENCE 28 (1993) 2531–2535

On non-Hookean behaviour of carbon fibres in bending

H. M. HAWTHORNE
Tribology and Mechanics Laboratory, National Research Council Canada, Vancouver, B.C., Canada V6S 2L2

The deformation characteristics of a range of single, large diameter pitch-based carbon fibres, and some PAN- and rayon-based fibres, were studied in elastica loop bending experiments. In pitch-based fibres, non-Hookean behaviour was found to occur at lower strains the greater the fibre anisotropy. Only elastic deformation to failure, at strains of 3%, was found for low-modulus, nearly isotropic pitch- and rayon-based fibres. The non-linear elastic behaviour in pitch-based fibres resulted in pronounced hysteresis and discontinuities in the stress–strain plots on both unloading and reloading the loops. These effects are due to the initiation, propagation and possible healing of transverse microcracks on the compression side of the fibres.

1. Introduction

Various aspects of the longitudinal compression mechanical behaviour of anisotropic polymer [1–4] and carbon [5–9] fibres, and their composites [10–13], have been reported. In particular, methods have been discussed for assessing the strain in carbon fibres when these are loaded in either bending or axial compression. The finding that Raman vibrational frequencies, as measured by Raman microscopy of single carbon fibre surfaces, are sensitive to strain [9] has renewed interest in this topic because of the potential application to carbon fibre composites. There is some evidence, however, that due to inherent band position scatter [14] the accuracy of this technique is limited to about 0.2% strain, which is a significant fraction of compressive failure strain in all but low-modulus carbon fibres [7, 15].

The compression (and tensile) deformation of single carbon fibres has been studied by elastica loop [5, 6] or similar knot flexure [8] tests, but recently these loop tests were described as problematic [9]. Elastica loop experiments were also successfully carried out by the writer some time ago on pitch-based carbon fibres (not mesophase pitch-based fibres), as well as on commercial fibres derived from both PAN and rayon. While reported in preliminary form [15], this work was never published in full. The recent publications prompt the present discussion of the results from these earlier experiments in the context of current work.

2. Experimental procedure

Specimens were mostly those obtained from the stretch-graphitization of pitch-based glassy carbon fibres. Thus, fibres were available with a range of physical and mechanical properties corresponding to microstructures from isotropic to highly axially oriented [16]. These were of considerably larger diameter than commercial PAN- and rayon-based fibres, which greatly facilitated both the single filament experiments and the observation of deformation effects.

Fibres were deformed in bending (flexure) using the simple elastica test [17] whereby a small fibre loop, immersed in light oil between glass slides, was loaded incrementally and the corresponding loop major and minor axis dimensions, ϕ and D, measured using a travelling microscope. Values of tensile or compressive stress and strain at the loop tip were calculated from these loop dimensions using the formulae [5]

$$\sigma = 60\, WD/\pi d^3 \qquad (1)$$

and

$$\varepsilon = 1.067\, d/D \qquad (2)$$

where σ is the maximum stress and ε is the maximum strain in the loop (at the tip) and the other parameters are defined in Fig. 1. These calculations are valid only so long as fibre deformation remains elastic and the loop thus retains its theoretical shape. Nevertheless, when non-elastic deformation was found, apparent stress and apparent strain values were derived similarly and used for data presentation purposes.

3. Results

Fig. 2 illustrates the two basic deformation responses observed in pitch-based fibres, to the increasingly severe bending imposed in the elastica loop tests. Low-modulus fibres exhibit perfectly elastic behaviour, the loop retaining the theoretical elastica shape, with a ϕ/D ratio of 1.34 [6], right up to fibre failure. In contrast, loops formed from high-modulus fibres clearly deviated at some stage from the elastica profile, becoming progressively more elongated with increasing tightening of the loop.

0022-2461

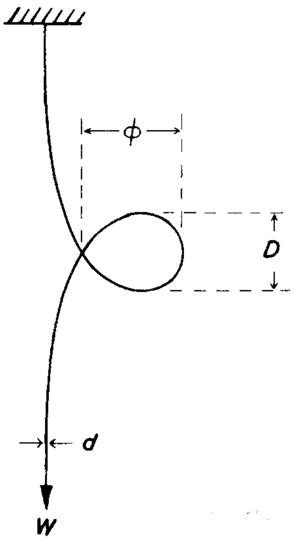

Figure 1 Schematic diagram of a fibre elastica loop.

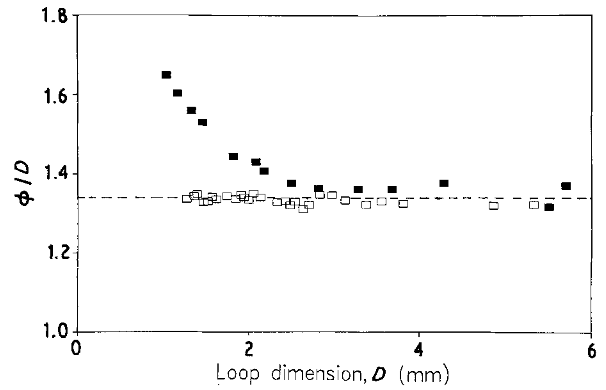

Figure 2 Loop dimensions characteristic of response of isotropic and anisotropic pitch-based carbon fibres to bending in the elastica test: (■) oriented fibre ($E = 260$ GPa, $d = 14$ μm); (□) isotropic fibre ($E = 35$ GPa, $d = 31$ μm); (---) theoretical elastica ratio.

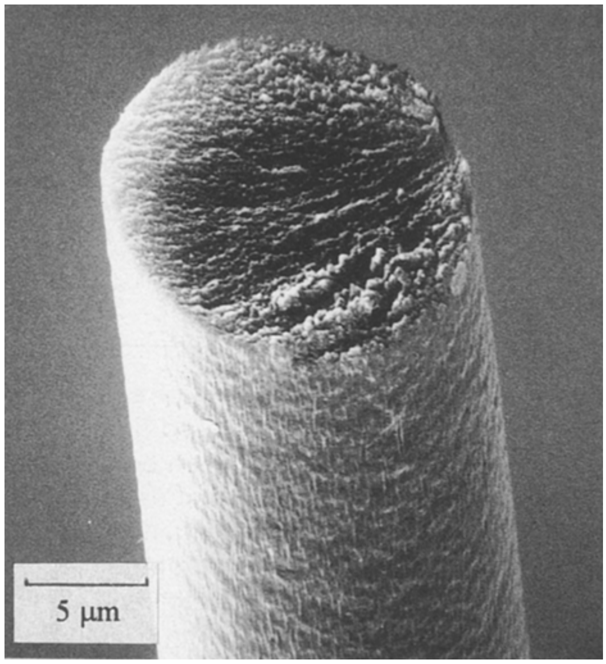

Figure 3 Tensile fracture surface of anisotropic pitch-based carbon fibres showing typical microstructure.

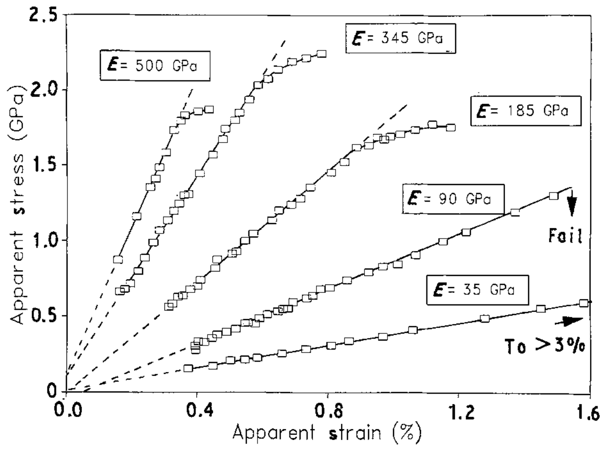

Figure 4 Plots of maximum stress and strain at the loop tip in elastica bending tests of various pitch-based carbon fibres.

Maximum stress and strain values at the loop tip could be calculated accurately in the case of these large and consistent diameter fibres. Fig. 3 shows a tensile failure surface of a high-modulus pitch-based fibre to illustrate the uniformly circular cross-section of these fibres. Also evident is the smoothness of the fibre surface compared to rayon- and PAN-based carbon fibres, and the densely packed and homogeneous nature of the fibre internal microstructure. The pitch-based carbon fibres have a similar onion-skin/random core structure to PAN-based fibres. In particular there are no large, folded, graphitic sheets or strong orientation textures as in mesophase pitch-based carbon fibres [18, 19].

Representative stress–strain plots from elastica loop experiments on a range of pitch-based carbon fibres of different Young's modulus are shown in Fig. 4. Only elastic deformation was found in low-modulus fibres, while non-linearity similar to that for other carbon fibres [5, 6] sets in with all filaments of Young's modulus greater than about 150 GPa, usually at lower strains the greater the modulus. This non-Hookean behaviour first appears in those present fibres which exhibit a degree of anisotropy, expressed as a ratio of tensile to shear modulus, close to that for PAN-based carbon fibres [6], as shown in Fig. 5.

Fig. 6 shows no differences in behaviour upon unloading and repeating the loading cycle of isotropic carbonized (1000 C) and "graphitized" (2800 C, unstretched) pitch-based fibres which had been loaded to high strains in the elastica loop. In contrast, hysteresis was observed in anisotropic fibres which had been strained in the loop beyond their linear elastic limit. Unloading and reloading such looped fibres produced stress–strain plots as in Fig. 7, showing first a reduction of the apparent modulus and then several jogged segments leading eventually to complete fibre fracture. Fibre unloading also occurs discontinuously. Although hysteresis effects in the bending of carbon fibres are known [5], this ratchet-like behaviour has not been reported before.

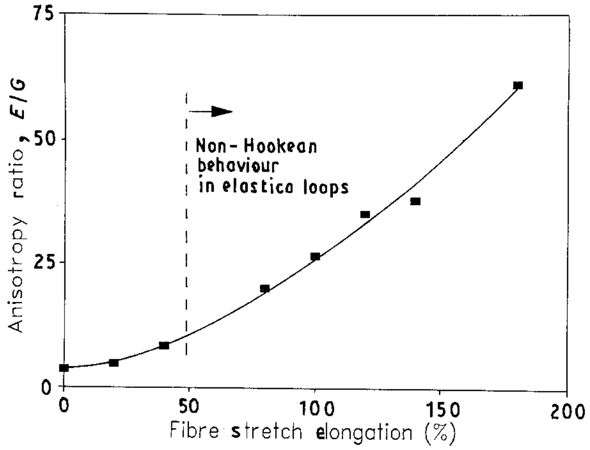

Figure 5 The anisotropy ratio and bending behaviour of various pitch-based carbon fibres.

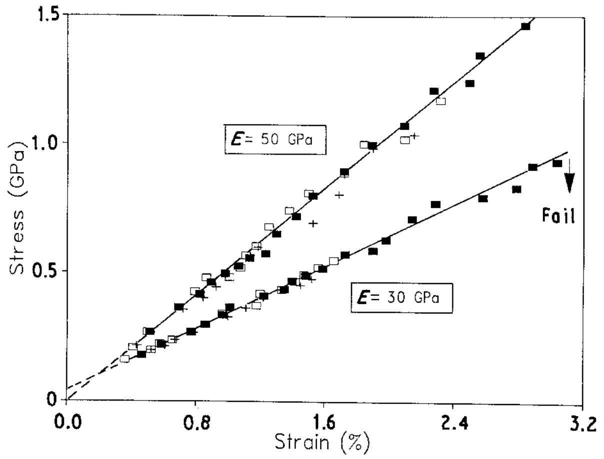

Figure 6 Elastica loop stress–strain plots for essentially isotropic pitch-based carbon fibres in repeat bending experiments: (□) first loading, (+) unloading, (■) second loading.

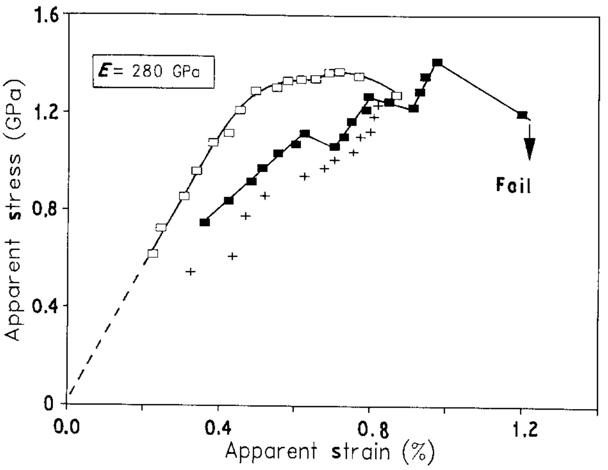

Figure 7 Elastica loop stress–strain plot typical of anisotropic, high-modulus pitch-based carbon fibres ($E = 280$ GPa) in repeat bending experiments: (□) first loading, (+) unloading, (■) second loading.

4. Discussion

Hysteresis effects in elastica loop tests [5] were reportedly found for all rayon-based fibres, irrespective of their degree of anisotropy. There are also differences in detail in the stress–strain plots for the anisotropic rayon-based [5] and pitch-based fibres. Furthermore, while a repeat of the loop experiments with Thornel VYB fibres gave similar low-strain results to those of Williams *et al.* [5], non-Hookean elastica effects were entirely absent since the ratio of loop diameters (ϕ/D) remained at the theoretical elastica value [6] up to failure at $> 3\%$ strain, as shown in Fig. 8. (An ill-defined shape of the initially very lightly stressed loops of these low-modulus, small-diameter fibres and possible rotation of the crenellated fibre to minimize its diameter in the bending plane [6] could explain the anomalous low-strain results.)

Non-Hookean behaviour occurs only in pitch-based carbon fibres which exhibit an anisotropy level close to that where it occurs in PAN-based fibres. Together with the absence of similar non-linear elastic deformation in very low modulus rayon-based fibres and the similar fracture strains from bending and axial compression tests, this suggested that the non-Hookean effects could have the same cause in all carbon fibres [7]. This was confirmed by SEM examination of strained loops, after the experiments. Transverse microcracks were observed, identical to those reported on PAN-based fibres [6], on the compression side of elastica loop tips formed by both anisotropic pitch- and rayon-based carbon fibres, as shown in Figs 9 and 10. No evidence was found for fibrillar destranding in high-modulus rayon-based fibres (Thornel 75), or compression microcracking in low-modulus fibres (Thornel VYB) at or near any of the highly strained loop tips examined.

Whether in axial compression or bending, transverse microcracking occurs under lower compressive strains the higher the preferred orientation, and hence axial modulus, in several types of carbon fibre [7, 15]. In addition to corroborating this, recent bending tests on PAN-based carbon fibres [9] showed such micro-

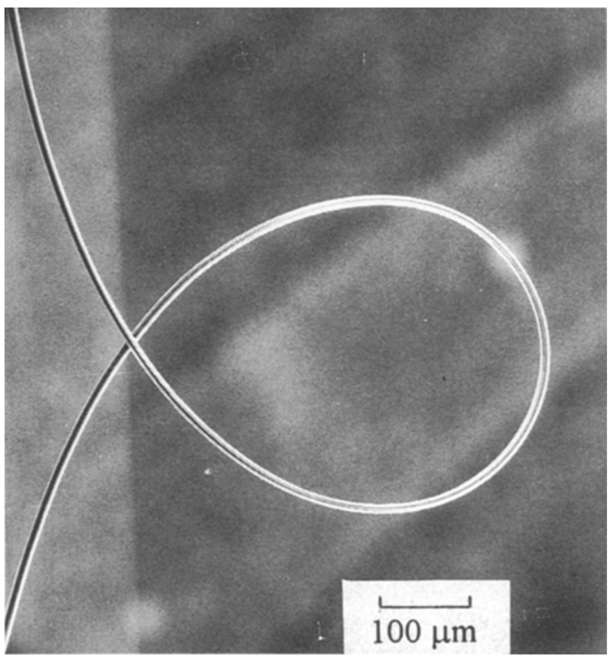

Figure 8 Low-modulus rayon-based (Thornel VYB) carbon fibre loop showing theoretical elastica shape ($\phi/D = 1.33$) even at 3.2% strain at loop tip.

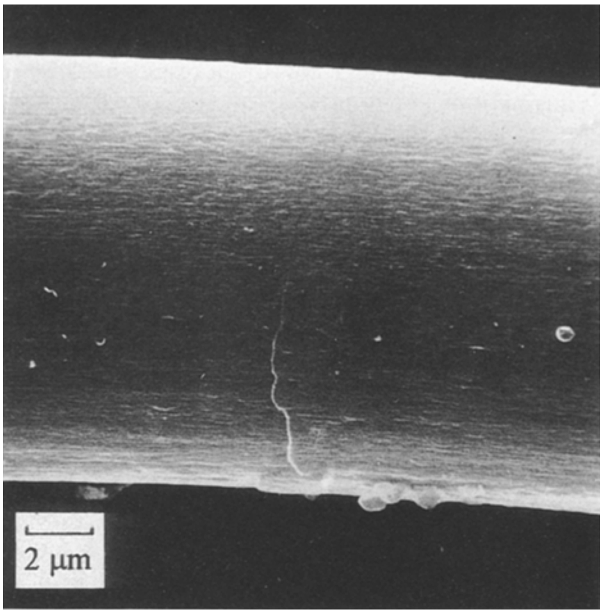

Figure 9 Typical transverse crack on compression side of anisotropic pitch-based carbon fibres bent beyond linear elastic region in loop tests.

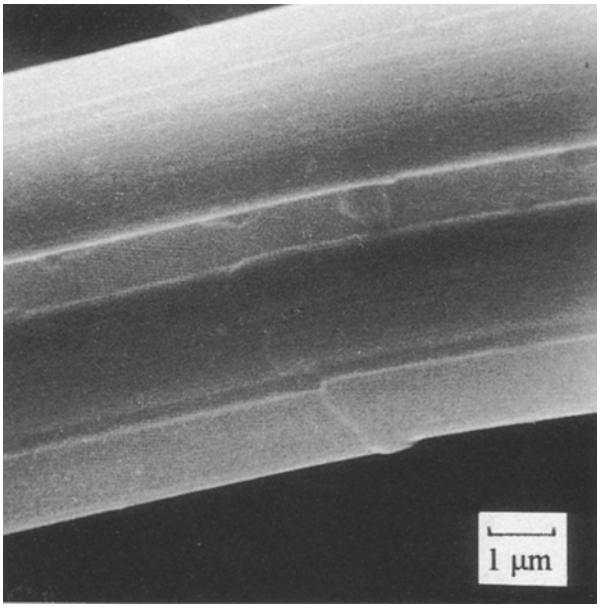

Figure 10 Typical transverse crack on compression side of anisotropic rayon-based carbon fibres bent beyond linear elastic region in loop tests.

cracks to be brittle fractures where fibre surface stress adjacent to the crack is substantially relieved by the fracture. Now the outer layers of most carbon fibres are more highly axially oriented than their core. For pitch-based fibres it was demonstrated that axial compression cracking occurs first in these more anisotropic surfaces [7] and it was suggested that this is also the case for cracks on the compression side of elastica loops [15]. The observed hysteresis effects thus result from the uncracked core and tensile side providing both a reduced restoring force on unloading the elastica loop and a lower effective modulus on initial reloading. Continued loop deformation proceeds by a combination of bending strain (the linear segments of the plot in Fig. 7) and either (or both) formation of additional cracks near the loop apex or a stepwise propagation of the first compression crack across the fibre surface and/or into the core (the short drops in the plot). The unloading steps may reflect an incremental crack healing process. The "extra cracks" explanation is the same as that given for the Raman frequency drops observed in bending experiments [9] which correspond to the first, rather than the second, loading cycle of the present tests. Eventually, however, complete fibre failure probably initiates by a Reynolds–Sharp mechanism [8, 20] on the tension side of the fibre where the strain increases rapidly regardless of how the compression strain is accommodated.

Although none were detected, the possibility of hysteresis effects in elastica loop bending of PAN-based carbon fibres was anticipated by Jones and Johnson [6]. Likewise, compression microcracks were not observed in knot flexure tests with PAN-based fibres [8]. In the latter case this may have been related to the coexistence of a significant torsional strain in the knots reducing the surface axial strain [8], or influencing the local fibre deformation response. However, the possibility remains that the small diameter of the fibres could have contributed to the experimental difficulties in both cases. As Raman microscopy can assess strain within small areas, it is suggested that a survey of surface strain distributions in both elastica loops and knots formed from various fibre types, coupled with subsequent embedding of the strained specimens and electron microscopic examination of sections, would be useful in further elucidating the details of carbon fibre deformation behaviour.

Finally, the deformation of fibres in torsion should be dominated by the characteristics of the material closest to the surface. Slope discontinuities in torsional stress–strain plots, in otherwise linear elastic behaviour to failure, were found for anisotropic pitch-based carbon fibres, very similar to those reported for PAN-based fibres [6]. Although undoubtedly arising from structural effects, observation of the torsional fracture surfaces revealed no sign of any type of cracking, sheath–core delamination or fibrillar de-stranding [5], in even the most highly oriented fibres. However, since compression cracking is now known to occur in high-modulus Thornel fibres, there is no need for the wire-rope analogy for plastic yielding involving separation and buckling of microfibrils within these fibres [5].

5. Conclusions

Only elastic deformation occurs in low-modulus pitch- and rayon-based carbon fibres in elastica loop bending tests. In contrast, in anisotropic pitch-based fibres non-Hookean bending initiates at lower strains the higher the fibre modulus, due to the onset of transverse microcracking on the compression side of the fibres. Subsequent hysteresis and discontinuous deformation effects in bending and relaxation of the highly strained loops are probably due to propagation and healing of these cracks.

References

1. J. H. GREENWOOD and P. G. ROSE, *J. Mater. Sci.* **9** (1974) 1809.
2. W. L. WU, V. F. HOLLAND and W. B. BLACK, *J. Mater. Sci. Lett.* **14** (1979) 250.
3. S. J. DeTERESA, R. S. PORTER and R. J. FARRIS, *J. Mater. Sci.* **23** (1988) 1886.
4. D. C. MARTIN and E. L. THOMAS, *ibid.* **26** (1991) 5171.
5. W. S. WILLIAMS, D. A. STEFFENS and R. BACON, *J. Appl. Phys.* **41** (1970) 4893.
6. W. R. JONES and J. W. JOHNSON, *Carbon* **9** (1971) 645.
7. H. M. HAWTHORNE and E. TEGHTSOONIAN, *J. Mater. Sci.* **10** (1975) 41.
8. J. L. G. DaSILVA and D. J. JOHNSON, *ibid.* **19** (1984) 3201.
9. N. MELANITIS and C. GALIOTIS, *ibid.* **25** (1990) 5081.
10. N. L. HANCOX, *ibid.* **10** (1975) 234.
11. H. T. HAHN and M. M. SOHI, *Compos. Sci. Technol.* **27** (1986) 25.
12. S. L. BAZHENOV and V. V. KOZEY, *J. Mater. Sci.* **26** (1991) 6764.
13. H. F. WU and J. R. YEH, *ibid.* **27** (1992) 755.
14. N. EVERALL and J. LUMSDON, *ibid.* **26** (1991) 5269.
15. H. M. HAWTHORNE and E. TEGHTSOONIAN, in Extended Abstracts of 12th Biennial Conference on Carbon, Pittsburgh, 1975 (American Carbon Society) p. 295.
16. H. M. HAWTHORNE, in Proceedings of 1st International Carbon Fibrés Conference, London, 1971, *Plastics & Polymers* Conference Supplement 5 (The Plastics Institute, London, 1971) p. 81.
17. D. SINCLAIR, *J. Appl. Phys.* **21** (1950) 380.
18. E. MORINOBU, *J. Mater. Sci.* **23** (1988) 598.
19. I. MOCHIDA, S. M. ZENG, Y. KORAI, T. HINO and H. TOSHIMA, *ibid.* **27** (1992) 1960.
20. W. N. REYNOLDS and J. V. SHARP, *Carbon* **12** (1974) 103.

Received 8 July
and accepted 4 August 1992