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An experimental study of end effects in the extensional deformation of polymers

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The sample geometry and the constraints of grips used in measurements of mechanical properties affect the values of the elastic constants obtained. This is known to be more serious for anisotropic materials. Three polymers have been examined in extension: oriented polyethylene terephthalate, isotropic low-density polyethylene, and oriented nylon 6. The results all show a linear increase of compliance with increasing reciprocal sample length. Subtracting the displacements of samples so short that the grips were initially in contact ("zero-length" samples) indicates that the effect is caused mainly by shear in the grips. Extrapolation of the results to zero reciprocal length gives the true sample compliance, and this removes discrepancies in comparing shear compliances derived from extensional and torsional methods.

1. Introduction

Measurements of the elastic constants of materials are always affected by constraints and other contributions from the apparatus used, unless special precautions are taken. Systematic errors from these sources become particularly important when comparing measurements made by different techniques, especially when these are combined to extract further elastic constants. The effects are due to some combination of the geometry of the sample used and of the effects of grips or other constraints in the apparatus. The sample dimensions affect the results whichever method of deformation is used. In extension and in torsion the grips will contribute either by sample constraint or damage (a local alteration of the properties of the sample between the grips, which may be felt outside them), and in simple shear by distortion of the ends [1] and by compressive forces needed to keep the samples in place during the tests [2]. Three-point and four-point bending techniques are freer from additional effects as distortion at the contact points is likely to be very small, but the results are harder to analyse owing to the non-uniform distribution of stress both along and across the sample. The three-point method also has a small contribution from shear which depends on sample dimensions.

The effects of the ends may propagate some distance into the sample, and Saint-Venant made an early study of this phenomenon which led to the principle named after him: end effects in extension or torsion of cylinders or in the bending of beams decay to negligible proportions within a length approximately equal to the maximum cross-sectional dimension. Horgan [3] has made a theoretical study of the problem of extension of anisotropic and transversely isotropic materials from the point of view of the decay of strain energy. He showed that the interior stresses decayed exponentially from the ends with a characteristic (upper-bound) decay length given by $\lambda \simeq h(E/G)^{1/2}$, where h is the maximum cross-sectional dimension, E is the Young's modulus in the test direction and G the shear modulus of the plane containing h and the test direction. For large anisotropy E/G will be large, and so λ will also be large. Arridge and Folkes [4] have used finite-element models in plane strain to test this theory, and have found good agreement for both isotropic and anisotropic samples.

So it can be seen that the problem of measuring true elastic constants in extension becomes most acute with very anisotropic materials such as oriented block copolymers and ultra-high modulus polyethylene. End effects propagating a long way

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into the samples have been found in the former by Arridge and Folkes [4] and in the latter by Arridge, *et al.* [5]. A similar propagation has been found in torsion of fibre composites by Folkes and Arridge [6]. Some small effects have been found in simple shear, which have been described elsewhere [2]. The problem may be tackled by finite-element methods by looking at the deformation of and within the grips themselves, but this is difficult, and an empirical approach would seem to be easier. This paper will deal with end effects found in extension of thin sheets of some polymers, and will suggest a way of eliminating them. The case of torsion will be discussed elsewhere [7].

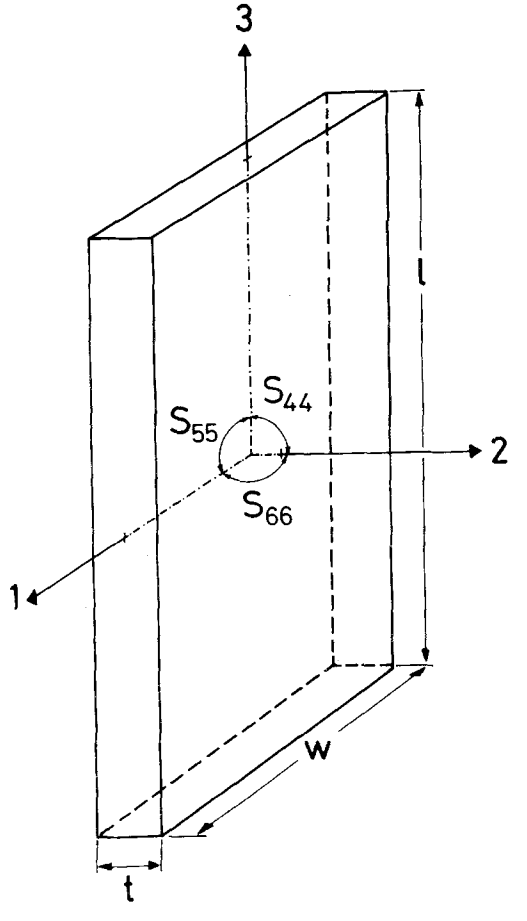

Figure 1 Diagram showing Cartesian axes with respect to sample sheet: 3 is parallel to the initial draw direction, 2 is normal to the sheet, and 1 is the transverse direction. The shear compliances S_{44}, S_{55} and S_{66} correspond to shear of the planes normal to the 1, 2 and 3 directions respectively. For a sample cut perpendicular to the draw direction, l and w will be interchanged.

2. Theoretical considerations

We shall be dealing with samples in the form of flat sheets. Conventionally, the draw direction is called 3, the normal to the sheet is 2, and the transverse direction 1; for samples cut from this sheet the length, width and thickness will be denoted by l, w and t respectively (Fig. 1). For anisotropic materials and small enough strains the elastic behaviour should obey the generalized Hooke's Law, which is given in matrix or engineering notation by

$$\epsilon_i = \sum_{j=1}^{6} S_{ij}\sigma_j, \qquad (1)$$

where ϵ_i are the strains, σ_j the stresses and S_{ij} the corresponding compliances. Values of j of 4, 5 or 6 refer to shear of the planes normal to 1, 2 or 3 respectively. For a sample of orthorhombic symmetry there are 9 independent, non-zero compliances: the extensional compliances S_{11}, S_{22}, S_{33}; the shear compliances S_{44}, S_{55}, S_{66}; and the off-diagonal components S_{12}, S_{13}, S_{23} of the compliance matrix which are related to Poisson's ratios ν_{ij} by the relation $\nu_{ij} = -S_{ij}/S_{jj}$. Since all the off-diagonal shear compliances are zero, the index j in extension in Equation 1 need run over values 1, 2 and 3 only.

In extensional tests we could imagine in principle that the end effects might arise because the grips constrain the sample to constant width, preventing lateral contraction locally. In this case the deformation would approach plane strain, and it can be shown that this would lead to an effective reduction of the extensional compliance (see Appendix). However, the tensile stress is transferred inwards from the grips via shear, and since it is concentrated in the surface layers at the ends, this leads to an apparent increase in the extensional compliance [4]. It is also possible that some plastic deformation may take place locally within the gripped region owing to very high local shear stresses. Polymers are strictly non-linear viscoelastic materials, and inhomogeneous stress fields can give rise to local stresses great enough to produce plastic deformation.

In spite of these possible complexities it seems adequate to suppose that any effects due to plastic deformation can be neglected, and that the problem may be considered as an elastic deformation in which there is a total end effect which decays away from the grips until the stress becomes uniform. Moreover, it will be shown that

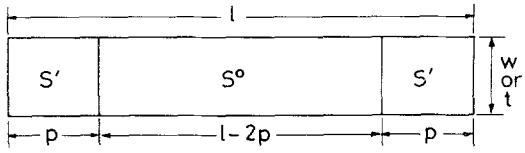

Figure 2 The three-block model of Folkes and Arridge [6] in terms of compliances. p is related to the characteristic decay length into the sample, and the other quantities are defined in the text.

the effects of the ends may be approximated by regarding the sample as consisting of a central unaffected region and a region at each end of length p with different properties, all in series. This is based on the three-block model of Folkes and Arridge [6] shown in Fig. 2. Here, p is related to the characteristic decay length, the primed quantities refer to the end regions, and those with superscript zero to the central part. Assuming uniform load σ along the sample, the total displacement is the sum of the displacements of the separate regions:

$$\sigma l S_{\text{eff}} = \sigma(l-2p)S^0 + 2\sigma p S', \qquad \text{for } l \geqslant 2p \quad (2)$$

which leads to the effective compliance

$$S_{\text{eff}} = S^0 + (2p/l)(S' - S^0) \qquad (l \geqslant 2p) \quad (3)$$

In these equations, p may be equated with Horgan's λ, so that the relevant expressions for p will be $t(S_{44}/S_{33})^{1/2}$ in the 3 direction or $t(S_{66}/S_{11})^{1/2}$ in the 1 direction. Plotting S_{eff} against $1/l$ should give a straight line starting from S^0 at infinite l and rising or falling to a value of S' at $l = 2p$ where the end regions merge, and thereafter $S_{\text{eff}} = S'$ and constant (Fig. 3).

3. Samples

Three different polymers have been examined. The first was biaxially-oriented polyethylene terephthalate (PET), extended at constant width to a draw ratio of 5. It was chosen because it is relatively easy to work with, having low time

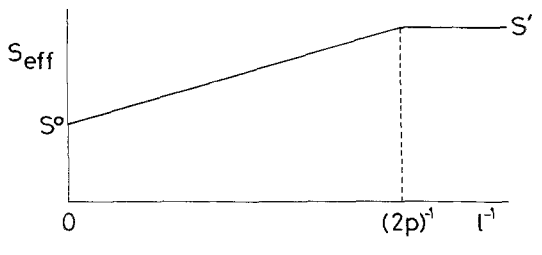

Figure 3 Expected behaviour of measured compliance S_{eff} as a function of inverse length l^{-1}.

TABLE I Compliances of 5:1 PET sheet from the work of Wilson ($S_{ij} \times 10^{10}$ m^2 N^{-1}) [9–12]

Extensional		Torsional		Off-diagonal	
S_{11}	4.00 ± 0.12	S_{44}	112 ± 5	S_{12}	−3.8 ± 0.4
S_{22}	9.0 ± 1.6	S_{55}	5.43 ± 0.23	S_{13}	−0.18 ± 0.01
S_{33}	0.77 ± 0.01	S_{66}	142 ± 7	S_{23}	−0.37 ± 0.05

dependences, is easily available, and its compliances have been measured [8–12] but without considering end effects. It is highly anisotropic and so should show up any effects well. The compliances are listed in Table I. The second polymer was isotropic low-density polyethylene (LDPE) which was prepared in our laboratories according to a standard procedure [2, 13–16] and has also been well characterized. Samples of this have been made with a thickness of up to 2 mm whereas the PET was limited to about 0.25 mm. The third polymer was the α phase of nylon 6 in both oriented and isotropic forms, annealed at 195° C and taken to the pure α phase by reaction with 3.5% aqueous phenol solution [17].

4. Experimental

The extensional compliances were all measured using a modified form of the dead-loading extensional creep apparatus of Gupta and Ward [13]. The position of the lower clamp was sensed by a Boulton Paul linear differential transformer transducer (type 53) leading to a Creep Monitor type D8 made by RDP Electronics, Ltd. By careful control of the hanging sample position with respect to the transducer coil former, repeatable readings to 0.1 μm (necessary for the shortest samples) could be taken directly from the precalibrated transducer meter. Larger displacements were measured by a null method using a micrometer head attached to the upper clamp.

The type of grips used were the standard design for flat samples as used in our laboratory, and shown diagrammatically in Fig. 4. They were capable of holding samples up to 3.2 mm wide and about 2.0 mm thick to a depth of 7.5 mm between stainless steel T-shaped plates T (about 0.6 mm thick) which helped distribute the compressive stress as uniformly across the gripped ends as possible. The grub screws G were tightened as nearly as possible to the same torque each time, although later the effect of screw tightness was found to be insignificant within the range of tightness used.

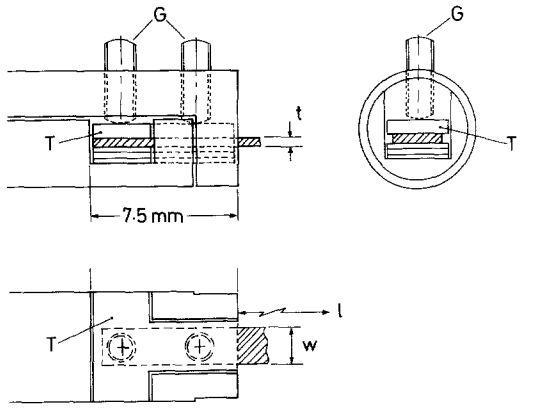

Figure 4 Sections through one of the grips used. The items G are grub screws pressing onto the T-shaped plate T which holds the sample (shaded) in place. The narrower, unmarked strips are spacers of the same shape as T used to ensure that samples of differing thickness are centred along the grip axis.

Displacements from apparatus distortion were measured using an effectively infinitely stiff sample clamped between the grips and measuring the displacements as a function of load. These were linear with load and were subtracted from all subsequent readings. The polymer samples themselves were first conditioned for reproducibility, and all readings were taken 10 sec after applying the load, waiting for recovery for 110 sec before applying the next load. Compliance–strain results were corrected to 20.0° C throughout and extrapolated to zero strain if the stress–strain behaviour was nonlinear. The nylon samples were conditioned to 65% relative humidity (r.h.), and these runs carried out in an environmental chamber maintained at 65% r.h. [17]. The resulting compliances were then all ten-second values at 20.0° C (and at zero strain where necessary).

5. Results

5.1. Polyethylene terephthalate

The PET samples were examined first. There were several of these: four cut parallel to the draw direction for S_{33} (two each of 1.2 mm and 2.5 mm width); one cut perpendicular to the draw direction for S_{11}; one cut at 45° for the compliance $S(45)$ (which is a function of S_{11}, S_{33}, S_{13} and S_{55}); and finally another parallel to the draw direction, but cut from near the edge of the original sheet of PET, where it was thicker, so that any thickness effect or property variation might be examined. The initial dimensions of all these samples are given in Table II. For each sample the compliance was measured at the full length, and then a piece was cut off and the compliance measured in the same way after again conditioning for reproducibility. This procedure was repeated until the length between the grips was around 4 mm; later runs went down to even shorter lengths. The upper grip was left untouched throughout each sample run.

The stress–strain behaviour was linear for all samples up to the maximum loads used (600 g). When the compliance was plotted against $1/l$, as suggested from Equation 3, straight lines were obtained (Figs. 5a to d). The slope of each is positive. This was to be expected as any contribution from plane strain would be negligible (the values of the expression $S_{13}^2/S_{11}S_{33}$ from the Appendix is only about 0.011 ± 0.001). Although at this time the minimum length exam-

TABLE II Dimensions of sample cross-sections and initial lengths between grips

Sample	Compliance		l (mm)	w (mm)	t (mm)
PET 5:1	S_{33}	(first narrow)	59.4	1.260 ± 0.010	0.251 ± 0.001
	S_{33}	(first wider)	94.3	2.427 ± 0.010	0.252 ± 0.001
	S_{33}	(second narrow)	34.2	1.184 ± 0.003	0.250 ± 0.001
	S_{33}	(second wider)	76.2	2.595 ± 0.009	0.247 ± 0.001
	S_{33}	(thicker)	55.6	2.028 ± 0.015	0.342 ± 0.001
	S_{11}		63.8	2.785 ± 0.024	0.266 ± 0.008
	$S(45)$		64.5	1.456 ± 0.015	0.268 ± 0.001
LDPE Isotropic S_{33} or D			54.4	2.362 ± 0.010	1.765 ± 0.004
Nylon 6 Oriented S_{11}			67.45	2.216 ± 0.013	0.259 ± 0.003
Isotropic S_{33} or D			82.2	1.862 ± 0.014	0.635 ± 0.004

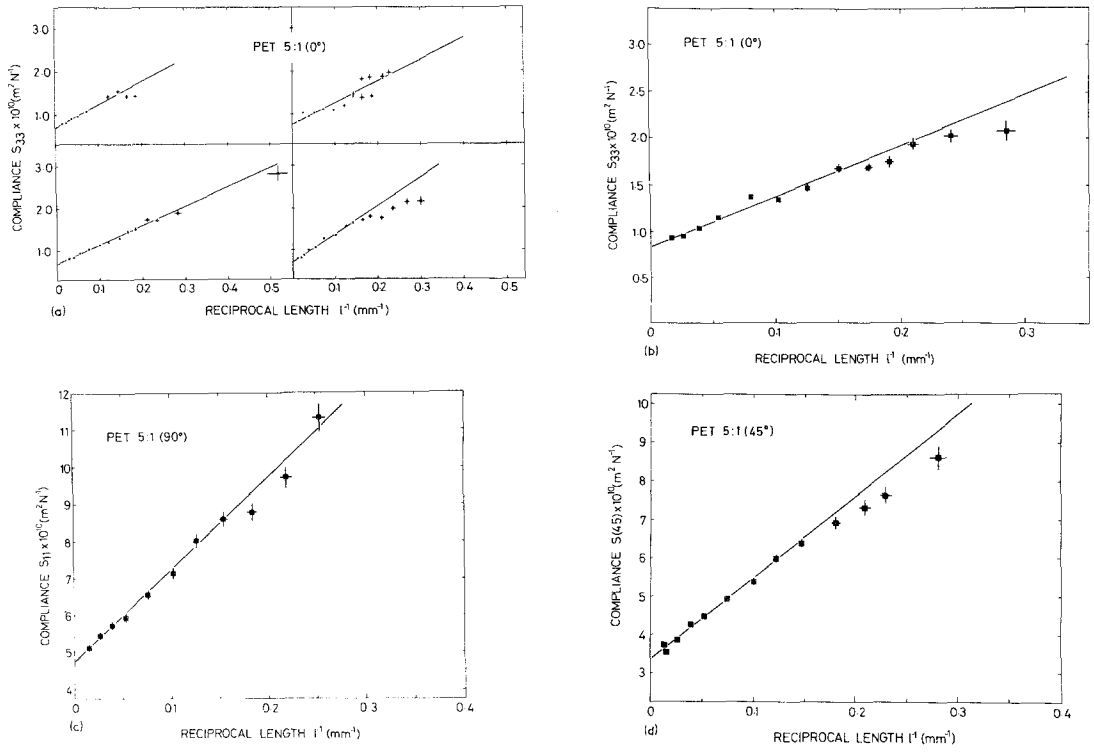

Figure 5 Extensional compliances versus reciprocal length for 5:1 drawn PET: (a) the four samples for S_{33} (parallel to the draw direction): the left-hand pair are the narrower samples ($w \sim 1.2$ mm) and the right-hand pair wider ($w \sim 2.5$ mm): the upper pair are the first set examined, the lower pair being examined later; (b) the thicker S_{33} sample ($t = 0.342$ mm); (c) the 90° sample for S_{11}; (d) the 45° sample for $S(45)$. Dimensions of all are given in Table II.

ined was about 4 mm, three of the S_{33} graphs show a tendency to level off near the value of $1/l$ given by the Horgan formula as adapted by Arridge and Folkes [4]. This gives $2p = 2\lambda = 2t(E/G)^{1/2}$ with E and G calculated appropriately from the orientation, i.e. $2t(S_{44}/S_{33})^{1/2} = 6.05$ mm, $1/l = 0.165\,\text{mm}^{-1}$ (for thicker sample 8.28 mm, $1/l = 0.121\,\text{mm}^{-1}$). The graph for S_{11} (Fig. 5c) does not level off, as here the expected length for this to be under way, $2p = 2t(S_{66}/S_{11})^{1/2}$, is about 2.8 mm and $1/l = 0.36\,\text{mm}^{-1}$. The graph for $S(45)$ (Fig. 5d) shows some tendency to level off. The evidence for a cut-off as predicted from the three-block model with propagation of end effects away from the grips is at best inconclusive, especially as two of the S_{33} graphs show no sign of levelling off even at the smallest lengths. Later on, more experiments were done to examine this further (Section 5.4).

5.2. Polyethylene

5.2.1. Isotropic low-density polyethylene

This sample was treated in the same way except that care had to be taken not to exceed the limit of stress–strain linearity, which was here 0.5% strain (cf. [14]). The initial dimensions are given in Table II and the results shown in Fig. 6. For an isotropic material in plane strain, Equation A2 becomes:

$$S_{33_{ps}} = S_{33}[1 - \nu_{13}^2], \qquad (4)$$

and since ν_{13} for this polymer is close to 0.5 [14, 15], $S_{33_{ps}} = 0.75 S_{33}$, an appreciable reduction. This indicates that plane strain may be contributing somewhat to the total effect.

5.2.2. Ultra-high modulus linear polyethylene

If the results for ultra-high modulus linear polyethylene of draw ratio 28 [5] are plotted in the same fashion, they appear as in Fig. 7. The end effects are very pronounced but the graph shows no straight-line sections; this is probably a result of the quite different gripping method used, so that the width and thickness effects would interact in a different way. There is likely to be very little con-

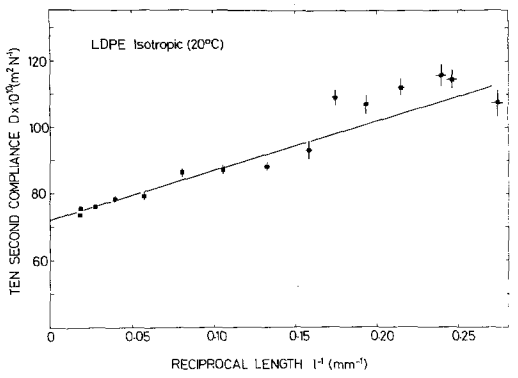

Figure 6 Extensional compliance versus reciprocal length for isotropic low-density polyethylene. The solid line is a weighted least-squares linear fit through the first nine points, and then extrapolated.

tribution from plane strain as the anisotropy was so high ($S_{33} \ll S_{11}$).

5.3. Nylon 6 at 65% r.h.

The third polymer examined was oriented nylon 6, which had been cold drawn to a ratio of 3.3, annealed at 195° C and taken to the pure α phase by phenol treatment [17]. The sample was cut out at right angles to the draw direction, so that S_{11} was being measured. This compliance is less dependent upon temperature and r.h. than S_{33}, and more linear in stress–strain behaviour [17]. The initial sample dimensions are listed in Table II. This time the results were taken to even shorter lengths than before, 1.1 mm, and later even one of "zero-length" where the sample was short enough for the grips to be initially in contact. The results were linear to 0.37% strain for longer samples but shorter ones (<4.5 mm) had to be extrapolated to zero strain. They are shown in Fig. 8 and again the slope is positive. With the value of $S_{66} = 5 \times 10^{-10}$ m^2 N^{-1} taken from earlier work [17] the expected cut-off would be around $2t(S_{66}/S_{11})^{1/2}$, $= 2 \times 0.259(5/3.21)^{1/2} = 0.65$ mm with $1/l = 1.55$ mm^{-1}, which is off the graph, yet some evidence of levelling off can be seen. This may be related to a contribution from plane strain which is more important in oriented nylon 6 than in oriented PET as drawn nylon 6 α phase is not very anisotropic in this r.h. range, (S_{33} is 3.65×10^{-10} m^2 N^{-1} [17]). The value for ν_{13} was taken to be that for nylon fibres, around 0.48 [18–20], which gave $S_{13} = -\nu_{13}S_{33} = -1.75$ and hence $S_{11_{\text{eff}}} = (0.74 \pm 0.03)S_{11}$ from Equation A3, a significant effect. The significance of the zero-length sample will now be discussed.

5.4. The zero-length samples

As a further attempt to gain insight into the mechanisms involved we looked at samples which were so short that the upper and lower grips were in contact initially; these were called "zero-length" samples. This was done for most of the samples examined and also for an unoriented sample of α nylon 6 (dimensions, Table II). For each sample small displacements were found upon loading which depended on the sample under test and were in the range 5 to 18 μm for a 500 g load (except for the thicker LDPE sample which gave 13 μm for 250 g). In all cases except the oriented nylon 6 the displacements were roughly linear with load. These displacements showed that there must have been some propagation of grip effects back into the gripped regions, as any effects due to the rest of the apparatus had been removed.

In order to see whether this effect was responsible for all of the end effects shown in the results for S versus $1/l$, the zero-length displacements were

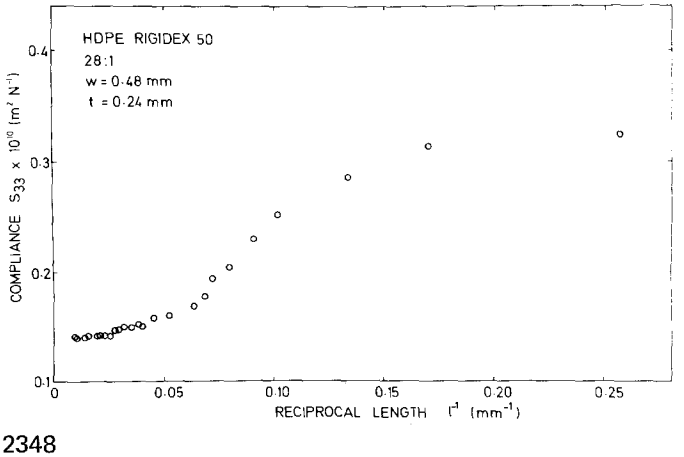

Figure 7 The results for ultra-high modulus linear polyethylene of draw ratio 28 [5] replotted as extensional compliance S_{33} versus reciprocal length.

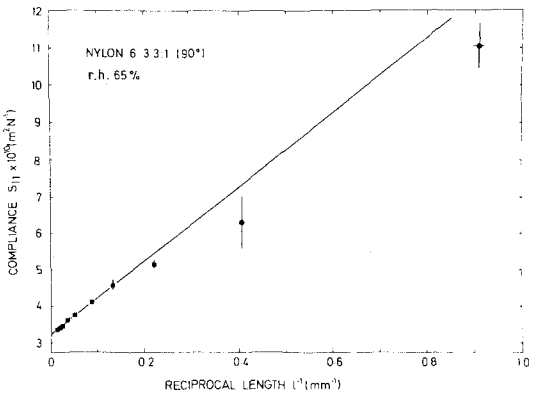

Figure 8 Extensional compliance at 90° to the draw direction (S_{11}) versus reciprocal length for 3.3:1 α phase nylon 6. All results at 65% r.h.

removed from the readings. This was done in two alternative ways: (1) subtracting the zero-length displacement from the measured displacement for each load and recalculating the results; (2) calculating what extra length δl the zero-length displacements were equivalent to, adding these to the measured lengths and recalculating. The results for δl are given in Table III. The two methods gave similar answers, as expected, and the corrected compliance values lay on straight lines of around zero gradient which were necessarily extrapolated back to the same infinite-length compliance value as before. The results are shown in Figs. 9 to 11. In the case of the more careful studies on oriented nylon 6 the two methods gave horizontal straight lines (χ_0^2 being 10 for the 10 points, showing an adequate fit) and the mean value of all the corrected points, $(3.24 \pm 0.02) \times 10^{-10}$ m^2 N^{-1}, was not significantly different from the original extrapolated value of $(3.21 \pm 0.04) \times 10^{-10}$ m^2 N^{-1}.

The isotropic nylon 6 sample was later used to check the effect of altering the compression stress distribution in the grips. This was done by repeating the zero-length experiment after removing the plate T of the lower grip; the mean displacement calculated for a standard tensile load equivalent to 10^5 N m^{-2} altered from 0.196 ± 0.009 μm to 0.176 ± 0.003 μm, a change of 2.1 σ, which is probably insignificant ($p \simeq 0.03$). The effect is not the grips themselves, and this is supported by the value of δl in all the experiments varying with the sample under test (Table III). So the end effect is mainly in the samples themselves.

In the oriented nylon the subtraction of zero-length displacements linearized the stress–strain relationship for all lengths. In the isotropic nylon the time dependence from 100 to 1000 sec $[(S(1000\,\text{sec}) - S(10\,\text{sec}))/S(10\,\text{sec})]$ had been checked and was reduced from 18.5% at 82.2 mm length to about 12% at zero length, implying that the creep properties of the gripped regions are different from those of the body of the sample.

6. Discussion and conclusions

In all the results obtained using the present grips, straight-line plots of compliance against reciprocal length have been obtained. This suggests a simple way of eliminating all end or grip effects. If the results are extrapolated back to $1/l = 0$ (infinite length), any effect will disappear and so systematic errors arising from them will vanish. The same technique has been applied to torsional compliance measurements [7] where the compliance decreases with decreasing sample length [6]; the $1/l \to 0$ extrapolation is still valid. When this is done, direct comparison of extensional and torsional compliances becomes possible.

In this way agreement has been improved in PET between S_{55} obtained from the $S(45)$ sample

TABLE III Extra lengths δl (mm) for zero-length samples

Sample	Compliance	w (mm)	t (mm)	δl (mm)
PET (5:1)	S_{33} (second narrow)	1.184	0.250	6.13 ± 0.33
	S_{33} (second wider)	2.595	0.247	7.56 ± 0.23
	S_{33} (thicker)	2.028	0.342	7.49 ± 0.34
	S_{11}	2.785	0.266	4.01 ± 0.11
	$S(45)$	1.456	0.268	4.41 ± 0.11
LDPE D (isotropic)	D (isotropic)	2.362	1.765	2.98 ± 0.06
Nylon 6 α form	S_{11} (3.3:1)			
	D (isotropic)	2.216	0.259	2.70 ± 0.10
		1.862	0.635	2.35 ± 0.13

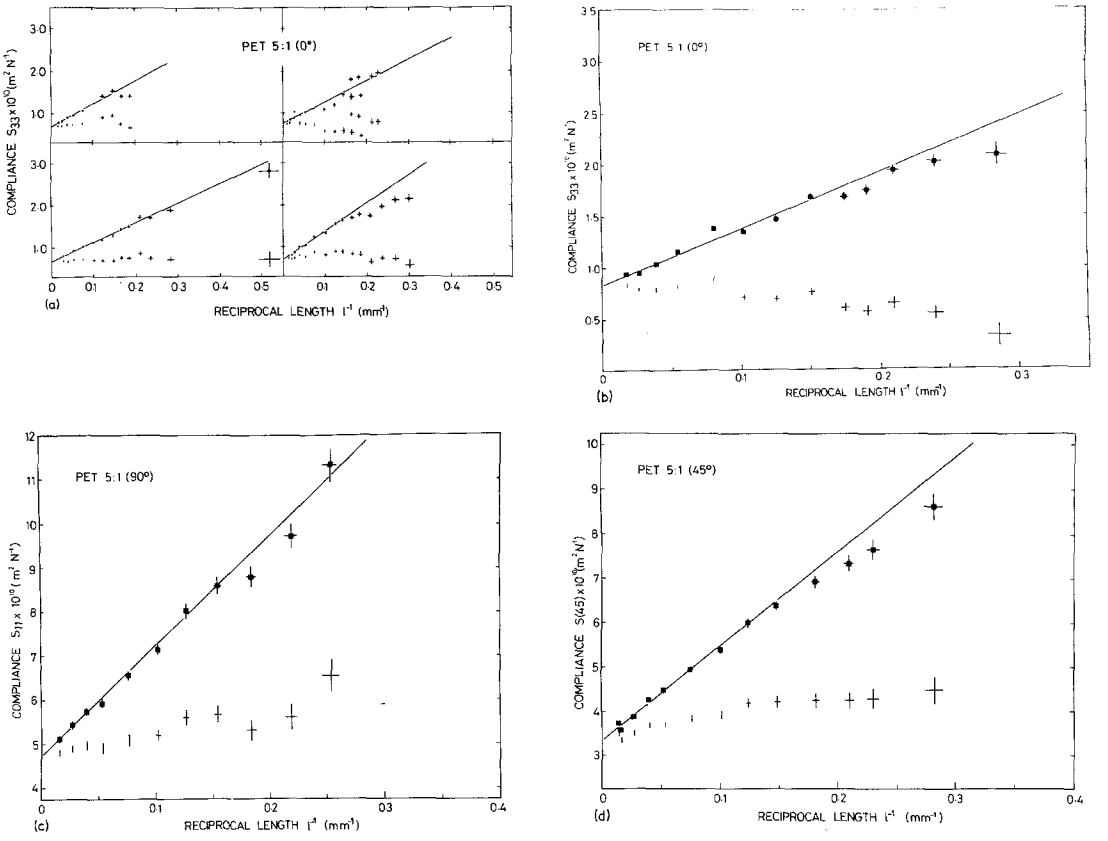

Figure 9 (a) to (d). All the results of Figs. 5a to d for PET with the points corrected for zero-length displacements added. These are plotted in plain vertical bars and crosses, and form almost horizontal straight lines.

using the relation

$$4S(45) = S_{11} + S_{33} + 2S_{13} + S_{55} \qquad (5)$$

which contains known compliances, and that obtained directly from torsion (called $\bar{S}_{55}$). Previously $\bar{S}_{55}$ has been significantly lower than S_{55} (e.g. [6]); Wilson has measured the compliances for PET samples similar to the present ones, using grips similar to the present ones [9–12, 7] and his results are listed in the left-hand side of Table IV. End effects had not been taken into account, and the difference between S_{55} and $\bar{S}_{55}$ is 1.84 ± 0.55 (in the units of Table IV), which is significant. Using the same relative variations with $1/l$ as in the present PET samples, correcting all these values to infinite length gives the set of figures in the right-

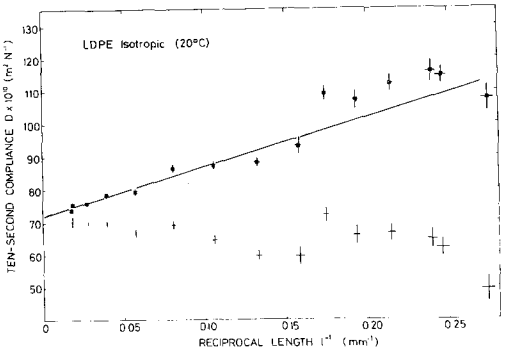

Figure 10 The results of Fig. 6 for isotropic low-density polyethylene with the points corrected for zero-length displacements added in plain bars and crosses.

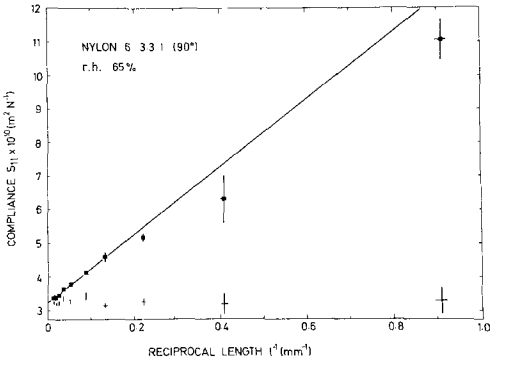

Figure 11 The results of Fig. 8 for nylon 6 with the points corrected for zero-length displacements added in plain bars and crosses.

TABLE IV Compliances for PET 5:1 sheet before and after correction for end effects ($S_{ij} \times 10^{10}$ m^2 N^{-1}). Torsional values for $\bar{S}_{55}$ from Lewis and Ward [7]

Compliance	Before correction	After correction
S_{11}	4.00 ± 0.12	3.61 ± 0.12
S_{33}	0.77 ± 0.01	0.66 ± 0.01
S_{13}	−0.18 ± 0.01	−0.18 ± 0.01
$S(45)$	2.92 ± 0.12	2.51 ± 0.12
S_{55} (calculated)	7.27 ± 0.50	6.13 ± 0.50
$\bar{S}_{55}$	5.43 ± 0.23	5.64 ± 0.25
Difference	1.84 ± 0.55	0.49 ± 0.56

hand side of Table IV; S_{13} has been assumed unchanged as the effect of mechanism A is negligible, and under the conditions this compliance was measured, using a fiducial grid [14, 10], end effects should be negligible. The difference is now 0.49 ± 0.56, which is insignificant. Thus the main source of disagreement between the two methods of measuring S_{55} is seen to be the end effects.

The effect of subtracting off the zero-length displacements gave almost horizontal straight lines in the graphs for compliance versus $1/l$ for all the samples (except perhaps the isotropic LDPE where it overcorrected somewhat). This implies that the major factor giving rise to the increasing compliance with shorter lengths in these samples is shear within the grips. It seems to be independent of total sample length, as the three-block model with continuity of stress gives straight lines for all these samples, except at shortest lengths where levelling off may start. Thus, for the present grips, the end "blocks" appear to be within the grips rather than propagating away from them. We may rewrite Equation 2 as

$$\sigma l S_{\text{eff}} = \sigma l S^0 + \sigma K_e, \tag{6}$$

where K_e is the length-independent extra displacements of the ends per unit stress. Then

$$S_{\text{eff}} = S^0 + K_e/l, \tag{7}$$

which gives the linear variation observed without bringing in the (outward) penetration length p. This means that within the linear range the true, end-free value S^0 could be obtained, in principle, from two samples of different lengths, to obtain the gradient K_e of the straight line. Clearly, this procedure should not be adopted, *a priori*, for any grip system or samples without a thorough study of the end effects similar to that outlined here.

For the isotropic LDPE the zero-length method seems to over-correct, but extrapolating results from other lengths to infinite length will still give the end-free compliance. Thus, it can be seen that the extrapolation procedure is valid for experiments of this kind. The procedure is simple and relatively quick. For other gripping techniques such as that of Arridge *et al.* [5] it may not be possible to find a straight-line plot, but empirical extrapolation will still give end-free elastic constants.

Appendix

When the extensional deformation approaches plane strain, one of the components ϵ_i of the strain matrix tends to zero. If a sample is loaded along the 3 direction, the corresponding strain will in general be

$$\epsilon_3 = S_{31}\sigma_1 + S_{32}\sigma_2 + S_{33}\sigma_3 \tag{A1}$$

from Equation 1. For a very long sample there will be no restriction to width and thickness reduction far enough from the grips so that $\sigma_1 = \sigma_2 = 0$ and the effective compliance is $\epsilon_3/\sigma_3 = S_{33}$. In plane strain, σ_2 is still zero but $\sigma_1 \neq 0$. Now, from Equation 1 we have $\epsilon_1 = 0 = S_{11}\sigma_1 + S_{12}\sigma_2 + S_{13}\sigma_3 = S_{11}\sigma_1 + S_{13}\sigma_3$, which leads to the effective compliance in the 3 direction:

$$S_{33_{\text{ps}}} = S_{33}\left[1 - \nu_{13}^2 \frac{S_{33}}{S_{11}}\right] = S_{33}\left[1 - \frac{S_{13}^2}{S_{11}S_{33}}\right]. \tag{A2}$$

A similar expression holds for extension in the 1 direction:

$$S_{11_{\text{ps}}} = S_{11}\left[1 - \nu_{31}^2 \frac{S_{11}}{S_{33}}\right] = S_{11}\left[1 - \frac{S_{13}^2}{S_{11}S_{33}}\right]. \tag{A3}$$

So the constrained part of the sample near the grips should be stiffened by the amount in the brackets.

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