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Torsion and normal force responses of glassy polymers in the sub-yield regime

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ABSTRACT

While there is a major body of literature relating the non-linear response of both elastomers and polymer melts with the molecular structure and topology of the chains, the same cannot be said of the non-linear response, especially that below the yield point, for polymer glasses. Here we take an approach that uses ideas of finite deformation elasticity and apply them to the isochronal response of glassy polymers in torsional deformations. We find that, while the torsional (shear) responses are similar for different molecular structures, there seems to be a strong effect of side group relaxations on the normal forces. The results are discussed in terms of the strain energy density function and the opportunities for future research.

1. Introduction

Alan N. Gent's fundamental approach to research over the years, required a deep understanding of the mechanics of rubbery materials and this lead him to insightful contributions relevant to theory and to the development of novel and creative experiments. From my view, his work to analyze the mechanics of rubber within the framework of finite elasticity [1–3] were inspirational even though they are not his most widely cited works. But the questions that he began in those papers and carried through in later work [4,5] concerning the form of the strain energy function of rubber networks and how to measure it [6], still remain with us today. But, it can also be said that the field of rubber elasticity is very mature and part of that success is certainly attributable to Gent's entire body of work.

Considering the original inspiration, then, much of my interest is in polymeric glasses and their non-linear viscoelastic behavior. In some ways, just as Gent used mechanics as his primary “tool” of choice in interrogating a range of problems in rubber, it has been one of my goals to investigate materials and, in the case of glassy polymers, have found that a similar approach can be very fruitful. But with the glassy state, unlike the case of finite elasticity of rubber, the continuum theories grew up at nearly the same time as the molecular network models, hence there was a considerable amount of feed-back between the two groups of researchers. Similarly, in polymer melts, there is a strong theoretical, molecular underpinning to the non-linear behavior through such models as the reptation theory of Doi and Edwards [7–11] and the molecular stress model of Wagner [12–14], in addition to the range of phenomenological constitutive models [15–21], that were developed over the years preceding the modern molecular models. In the case of the mechanics of glasses, the situation is not so well developed: there seems to be little molecular theory to support an in depth understanding of the non-linear viscoelastic response of glassy polymers, especially in the sub-yield regime. In what follows, we focus on this regime and show that several interesting features of the non-linear behavior of glassy polymers are observed when the response is considered within a similar framework to what one might use in finite elasticity evaluations of materials. Specifically, we will examine the torsion experiment in which the sample is held at constant length by a normal force while a twist is applied.

2. Background

The problem of finite torsion of a cylinder held at constant length L was solved by Rivlin and Saunders [22] in 1951 and the solution is given by Eqs. (1) and (2) which refer to Fig. 1 for the geometries of relevance. If we twist the cylinder by an amount $\Psi = \Theta/L$, the torque T and normal force N can be determined in terms of the cylinder radius R and the derivatives of the strain energy density function W with respect to the invariants I_i of the left Cauchy–Green deformation tensor B and its inverse B^{-1} as follows:

$$T = \pi\Psi \int_0^R \left[\frac{\partial W}{\partial I_1} + \frac{\partial W}{\partial I_2}\right] r^3\, dr \tag{1}$$

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$$N = -2\pi\Psi^2 \int_0^R \left[\frac{\partial W}{\partial I_1} + 2\frac{\partial W}{\partial I_2}\right] r^3\, dr \qquad (2)$$

In the case of shearing deformations such as torsion, then the invariants of the deformation tensor are

$$I_1 = I_2 = 3 + \gamma^2 = 3 + \Psi^2 r^2 \qquad (3)$$

Of interest in looking at Eqs. (1) and (2) is the observation that, first, the torque is an odd function of the deformation and the normal force is an even function, i.e., they are first and second order effects, respectively. More importantly, we also see that the torque depends on the sum of W_1+W_2 while the normal force depends on the sum of W_1+2W_2, where we now refer to $W_i = \partial W/I_i$ for the strain energy function derivatives. It is this fact that leads to the framework in which we find that torsional experiments provide a rich field to investigate the nonlinear response of glassy polymers as described below. This framework is commonly used in melt and solution rheology in polymers but with the strain energy function derivatives being substituted, as appropriate, by the first and second normal stress differences. We do not look into melts and solutions further, but move on to examine the actual behavior of the glassy polymer in the torsional experiment. Because of the dearth of theory of the strain energy density function for these materials, what is to be described next is the observed phenomenology and some interpretation in terms of the strain energy density function derivatives as seen in Eqs. (1) and (2).

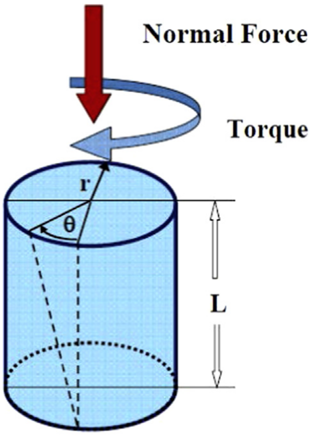

Fig. 1. Schematic of torsion of a cylinder held at constant length *L*.

3. Observations of the normal force magnitudes

As indicated above, while polymers are well known for their normal force effects in the melt state [23–31] and in cross-linked rubber [19,22,32–34], considerably less is known about the response of glassy polymers. And, although such normal stress or normal deformation effects in solids have been known for over a century since Poynting [35] first twisted wires, and others have cataloged ratcheting in plastic cycling of metal rods [36–38], to our knowledge the normal force response in a glassy polymer was not measured until the late 1970s [39,40]. When we did the experiments, not knowing what to expect, we simply made the measurements by taking tubes of a glassy poly(methyl methacrylate) and twisting them to strains of about 10% based on the outer tube diameter. The tubes were 3.33 cm in diameter with inner diameter of 2.54 cm. Fig. 2 shows [39,41], the nominal wall shear stress and the nominal wall normal stresses plotted against the logarithm of strain γ or the square of the strain γ^2. Because PMMA is viscoelastic, the values are isochrones (indicated in the figures) from stress relaxation measurements. Of interest is to back calculate the magnitudes of the normal forces for the PMMA. We find that for the 1 s isochrones and at a strain of 0.10 ($\gamma^2 = 0.01$) the normal stress is approximately 5 MPa and this would correspond to a normal force of approximately 1800 N. In retrospect this was quite large and shorter times gave even larger normal forces. Regardless, at that time we were interested in viscoelastic response of materials and constitutive modeling and not per se the magnitudes of the normal stresses. It is worth noting however, and something that lead to our subsequent thinking, that shortly after the work on PMMA, we tried to do similar experiments with polycarbonate (PC). The sample geometries were somewhat different, but the end result was that we did not succeed in the set of experiments because the normal forces were too low to be measured with sufficient accuracy for our purposes and we did not pursue the problem. This was in 1979–1980.

Nearly 20 years later [42], we carried out torsional measurements on 4 mm diameter solid cylinders of a polycarbonate material and the results are shown in Fig. 3 as double logarithmic representations of torque and normal force vs. the strain γ at the outer radius of the cylinder. Here the data show the expected linearity for the torque and the second order dependence of the normal force on the strain, both at small strain. As strain increases, the response begins to deviate as one might expect from straight line behavior. Of interest is that the maximum normal force of

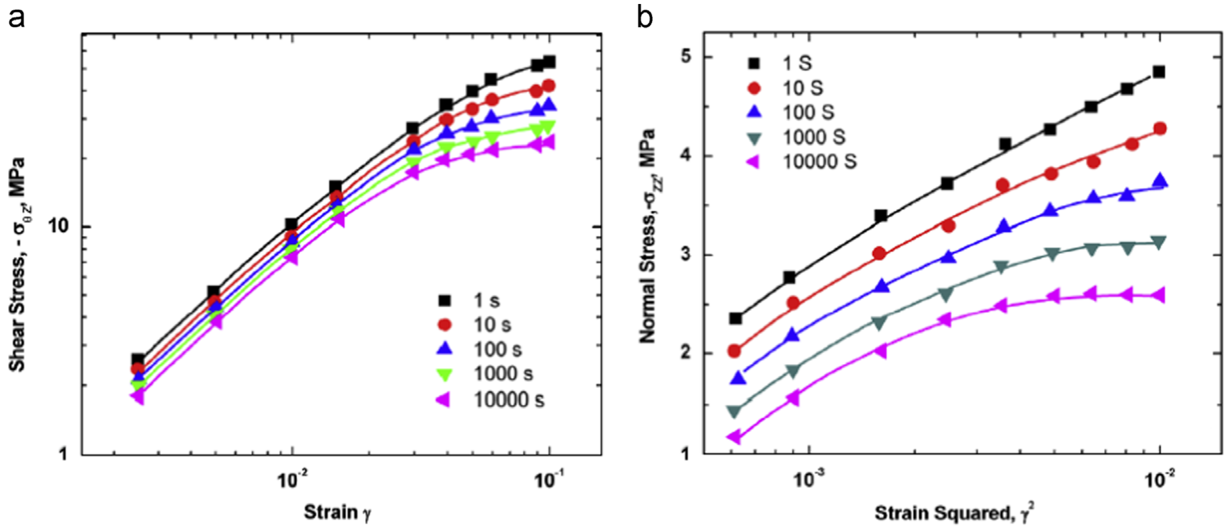

Fig. 2. Torsional data on a hollow cylinder of poly(methyl methacrylate) from stress relaxation isochrones. Data from McKenna and Zapas [39], plots from McKenna [41].