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Comparison between flexural and uniaxial compression tests to measure the elastic modulus of silica aerogel

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ABSTRACT

The Young's moduli of a set of silica aerogels have been measured by two techniques: 3-point bending and uniaxial compression. The data found by the two methods differ strongly. The uniaxial compression test gives generally underestimated values of Young's modulus, because of geometrical effects. The appropriate gauge lengths were estimated based on the discussion of Euler buckling and nonuniform stress distribution. The measured compressive moduli were analyzed to correct for machine compliance and possible misalignment under compression of the aerogels. Similarly, moduli obtained by 3-point bending depend on the length/thickness ratio of the sample, reaching equilibrium only for ratios above about 10. The corrected compressive moduli were comparable to those measured by 3-point bending on samples of sufficient length.

1. Introduction

The elastic properties of aerogels have been extensively studied, because of their importance for processing and applications of aerogels; moreover, aerogels are valuable as model materials for testing theories that relate network structure to properties. In this paper we demonstrate that experimental artifacts can produce misleading results regarding the dependence of elastic modulus on density. We show how to correct for these problems, so that the same results are obtained by different methods of measurement, such as uniaxial compression and 3-point bending.

Gel formation has been discussed in terms of percolation theory [1–5] and cluster–cluster or monomer–cluster growth processes [6,7]. The analogy between gel formation and percolation theory is based on experimental power-law evolution of physical properties, such as elastic modulus [8–10]. Another structural model based on the bending of cubic cells (proposed by Gibson and Ashby [11,12]) tries to describe the mechanical behavior of the porous open cells network and concludes that the Young's modulus, E, is proportional to the square of the bulk density, ρ.

To test the applicability of these models (percolation, growth process, structural model), the mechanical properties of silica alcogels and aerogels have been studied [4,5,13–15]. The results obtained on different sets of aerogels (by beam bending and ultrasound velocity measurements) show that the exponent of the power-law dependence of E on ρ is close to 3.8 [4,5]. The same value was obtained from finite element analysis of gel structures created by computer simulations of diffusion-limited aggregation [16–18].

In one study [19], a different exponent (2.85) was found, which the authors attributed to the peculiar microstructure of their aerogels. It must be noted that not only the kind of aerogel is different compared to the previous studies [4,5], but also the kind of test used to measure E, which was uniaxial compression. It is perfectly reasonable to imagine that the network structure will affect the exponent; however, before we can have confidence in such an interpretation, it is necessary to insure that the measurement technique provides reliable results.

Aerogels can be irreversibly deformed when subjected to isostatic pressure [20–22]. During the compression of the solid parts, new siloxane bonds are formed between aggregates by polycondensation reactions of silanol bonds [22]. This new reticulation between aggregates partially 'freezes' the network, increases the connectivity and, thus, the elastic rigidity. Therefore, it is essential that the modulus measurements be made using sufficiently small strains.

It has been shown [23,24] that the uniaxial compression test is sensitive to several technical or geometrical parameters that affect the measurements. One of the most important parameters is the ratio L_0/D [25–27] where L_0 and D are, respectively, the initial sample length and diameter. The uniaxial compression test is valid if this ratio ranges between two limits [28,29], corresponding to an

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inhomogeneous stress distribution for the lower limit and to the buckling effect for the upper limit.

The goal of this work is to check whether the different experimental results found in the literature on aerogels could result from artifacts in the mechanical test used. For this purpose, the elastic behavior of a single aerogel set will be characterized by the two methods previously cited: 3-point bending and uniaxial compression.

2. Experimental

2.1. Gel synthesis

Alcogels were made by hydrolysis and polycondensation reactions of tetramethoxysilane (TMOS) in ethanol. Hydrolysis was done with 4 moles of water per mole of TMOS. The gelling solution was poured into cylindrical tubes (diameter, $D = 6$ mm) and aged 2 weeks at room temperature. After aging, the gels were supercritically dried in an autoclave as previously described [5]. It is possible to tailor the bulk density of the aerogels by an adjustment of the TMOS concentration in the initial solution. By this procedure monolithic aerogels were made with densities ranging from 0.09 to 0.25 g/cm^3.

2.2. Mechanical tests

2.2.1. 3-Point bending test

For brittle material, the 3-point bending test is well suited to measure the Young's modulus in the static mode. A force P is applied by the displacement (δ) of the pushrod with a deflection rate equal to 500 μm/min; the distance between the two supports under the sample is called the span. A previous study [30] has shown the influence of the ratio span (s) to sample diameter (D) on the experimental results. If the ratio s/D is too small, the 'measured elastic modulus' is a combination of the Young's modulus and the shear modulus. A ratio higher than 10 is required to correctly measure the Young's modulus. Consequently, we have performed the bending (flexure) test with $s = 70$ mm and $s/D \approx 12$. The Young's modulus (denoted E_F) is calculated from the following relation:

$$E_F = \frac{4Ps^3}{3\pi D^4 \delta} \qquad (1)$$

The estimated error in E_F is 3%, calculated from the error estimates on P (10^{-5}), s (6×10^{-3}), D (3×10^{-3}) and δ (1×10^{-3}).

Indentation occurs at the points of contact with the pushrod and supports and must be taken into account to get an accurate measure of the elastic modulus [31] (see appendix). For the geometrical conditions used in this work, this effect is not significant, as the error in E_F from indentation is <10%.

2.2.2. Uniaxial compression

Uniaxial compression tests have been done using a RHEO-TAXT2/25/SMS machine. The accuracy of the uniaxial compression test [32] depends on the planarity and the parallelism of the contact area. Planarity and parallelism of the two metallic platens that compress the samples have been checked by a micrometric sensor (±0.1 μm), which measures the distance between the two flats. The measurements made at different points on the flats show that this distance is constant.

Great care was taken to ensure that the end faces of the test specimens were smooth and plane-parallel. The parallelism of the two contact areas of the aerogels is directly dependent on the sawing conditions. Precise control of the sawing procedure was achieved by means of a special fixture to hold the sample. Nevertheless, the stress–deformation curves show a short nonlinear evolution that can be attributed to the effect of the experimental set-up (defect in parallelism and planarity) (Fig. 1). In any case, the major part of the curve is clearly linear which proves the good proportionality between the stress and the deformation.

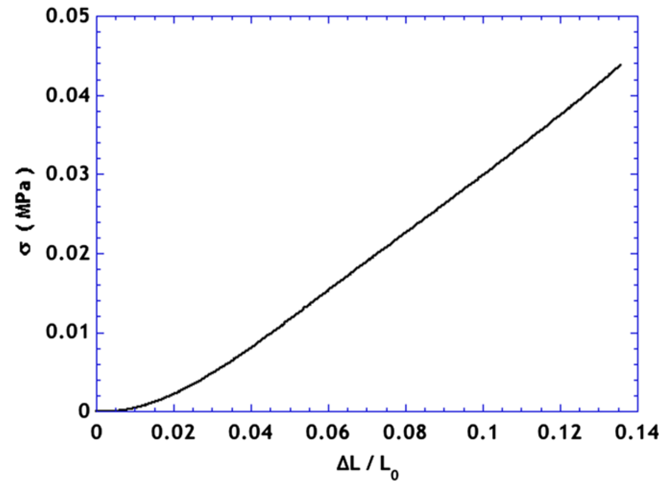

Fig. 1. Stress (σ) versus strain ($\Delta L/L_0$) showing reversible deformation of aerogel with density $\rho = 0.09$ g/cm^3 under uniaxial compression.

As we will show in Part 3, the geometrical conditions of the test are important. The ratio between the initial length L_0 and the diameter D of the sample is a pertinent parameter. The standard compression test specimen is a cylinder having an L_0/D ratio of 2 [19,23,24].

The Young's modulus (denoted E_C) measured by this test is calculated in the linear part of the curve by the following expression [32]:

$$E_c = \frac{4P}{\pi D^2 \ln(L/L_0)} \qquad (2)$$

where L = the sample length and L_0 the initial length. The error estimate is <1% calculated from the error estimates on P (10^{-5}), D (3×10^{-3}), and L (1×10^{-3}).

3. Results

Figs. 1 and 2 show that the sample behaves reversibly during a uniaxial compression test (Fig. 1), if the stress applied is not too

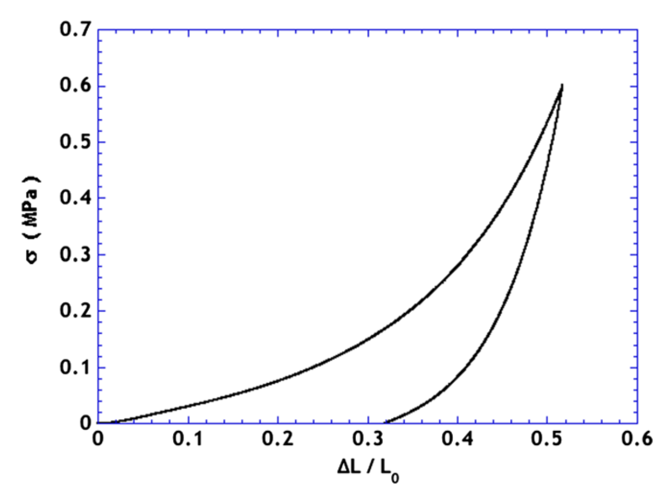

Fig. 2. Stress (σ) versus strain ($\Delta L/L_0$) showing irreversible deformation of aerogel with density $\rho = 0.09$ g/cm^3 under uniaxial compression.