DETERMINING THE ELASTIC MODULI OF REVERSIBLE GELS USING THE THEORY OF ELASTICITY

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INTRODUCTION

In our investigation of the properties of DNA-crosslinked polyacrylamide gels [1], there is a need to measure the elastic moduli of small (25μ l) samples of material. A static, nondestructive method that eliminates much of the material preparation and handling issues associated with traditional compressive tests has been developed.

METHOD

In the method, a rigid magnetic sphere is first embedded in the gel sample. An electromagnet is used to apply a force of known magnitude to the sphere, and the resulting deflection is measured after allowing for stress-relaxation to occur. The elastic modulus is determined from the force and displacement using the theory of linear elasticity [2]. The test fixture is shown in Figure 1a.

THEORY

Cylindrical coordinates (r, θ, z) are suitable for this problem of a rigid sphere embedded in an elastic medium of either infinite size or finite spherical volume (Figure 1b). In describing the geometry of the rigid sphere of radius R_0 , the radial spherical coordinate R is used. With the force applied in the z-direction, axial symmetry dictates that only radial and z displacements $(u_r \text{ and } w, \text{ respectively})$ are possible. Likewise, the only nonzero stresses expressed in common engineering notation are σ_r , σ_{θ} , σ_z , and τ_{rz} .

Case 1: Infinite Medium Approximation

The general solution to the irrotational deformation of an axisymmetric body can be obtained as a superposition of two strain fields, one generated from a harmonic strain potential, the other from a biharmonic displacement potential [2]. Using spherical harmonics to generate the potentials yields one such solution expressed by the displacement field:

$$u_r = \frac{Arz}{2GR^3} \left(1 - \frac{R_o}{R^2} \right) \tag{2}$$



Figure 1. (a) Photograph of test fixture. (b) Rigid sphere embedded in a spherical medium of radius R_1 and acted on by a force *F*.

$$w = \frac{A}{2G} \left[\frac{3 - 4\nu}{R} + \frac{R_o^2}{3R^3} + \frac{z^2}{R^3} \left(1 - \frac{R_o^2}{R^2} \right) \right]$$
(3)

In the displacement field, *A* is a constant to be determined from the boundary conditions, *G* is the shear modulus of the medium, and v is Poisson's ratio of the medium. The boundary conditions preclude the existence of radial displacements on the surface of the sphere and limit the *z*-displacement to a constant value, δ . This gives:

$$A = -\frac{3\delta R_o G}{-5+6\nu} \tag{4}$$

The stresses follow from the kinematic and stress-strain relations. Integrating the components acting in the *z*-direction over the surface of the sphere yields the net vertical force, or the applied force F:

$$F = \frac{24\pi\delta GR_o(-1+\nu)}{-5+6\nu} \tag{7}$$

For an incompressible material, the elastic modulus is:

$$E_i = \frac{F}{2\pi\delta R_o} \tag{8}$$

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Case 2: Finite Spherical Medium

For a spherical medium of radius R_1 , the solution can be obtained from the strain fields generated from the following set of harmonic (ζ) and biharmonic (Ψ) potentials:

$$\varsigma = A_1 R + A_2 \left(r^2 z^2 - \frac{1}{4} r^4 \right)$$
(9)

$$\Psi = \frac{A_3 z}{R^3} + A_4 z + \frac{1}{2} A_5 (5z^3 - 3zR^2)$$
(10)

The additional boundary condition that all displacements vanish at $R = R_1$ must then be imposed to determine the constants A_1 through A_5 . Proceeding as in the infinite medium case, the stresses in the *z*-direction can be obtained from the displacement field and then integrated to obtain the force. For incompressible media:

$$E_f = \frac{F}{2\pi\delta R_o} \frac{4n^5 - 5n^4 - 5n^3 + 5n^2 + 5n - 4}{4(n^5 + n^4 + n^3 + n^2 + n)}$$
(11)

The variable $n = R_i/R_o$ represents the size of the medium in relation to the embedded sphere. The error incurred by assuming an infinite medium can be expressed by the ratio of the elastic moduli:

$$\frac{E_i}{E_f} = \frac{4n^5 - 5n^4 - 5n^3 + 5n^2 + 5n - 4}{4(n^5 + n^4 + n^3 + n^2 + n)}$$
(12)

RESULTS AND DISCUSSION

Appropriateness of using the infinite medium approximation in determining the elastic modulus of the medium is dependent on the size of the medium in relation to the size of the sphere, as shown by Equation (12) and plotted in Figure 2. The error is approximately 25% at a relative size of n = 11, and decreases to 5% at n = 47.

The spherical medium is an idealized geometry chosen for the sake of simplicity. Most gel samples are held in containers that result in the presence a free boundary surface. Such conditions obviously become important as the size of the medium approaches that of the sphere. It is possible to determine the correction factor by modifying the elasticity problem to incorporate unique boundary conditions, including the case of slip between the sphere and the medium. However, if a second method of measuring stiffness (e.g. direct compression of a cylindrical sample) can be employed, the correction factor can be empirically derived.

In the DNA-crosslinked gels contained in conical, open-top vials, direct compression tests yielded a correction factor of 1.03. Figure 3 presents the elastic modulus as a function of relative crosslink density, where 100% crosslinking defines the maximum number of crosslinks permitted by the amount of DNA functional groups in the polyacrylamide chains [1]. The low stiffness of the gels is attributed to low concentrations of both monomer (3 mg per 100 ml of gel) and crosslinker (stoichiometrically equivalent to 0.28% by weight of the standard crosslinker bis-acrylamide). Based on the composition of bis-crosslinked gels, the monomer concentration can be increased at least ten-fold and the crosslink density by at least twenty-fold.

Equation 7 indicates that the value of Poisson's ratio does not significantly affect the value of the elastic modulus. Compared to the case of incompressibility, an illogical assumption of v = 0 reduces the elastic modulus by approximately 17%. Since typical values for the Poisson's ratio of elastomers are between 0.49 and 0.499 [3], the error associated with the assumption of incompressibility is negligible.

Particularly for thermoreversible gels, this method allows repeated testing of small samples. The sample is heated above its melting point, and a magnetic bead is positioned at the center of the gel. Upon gelation, the sample can be tested at various loads. A number of methods can be used to accurately measure the displacement of the sphere. At the completion of testing, the sphere can be removed, and the sample reused.

SUMMARY

A method has been devised that offers several advantages over traditional means of measuring the elastic moduli of soft materials. This method, based on the theory of linear elasticity, permits small samples to be tested without damage. Furthermore, sample preparation and handling can be greatly simplified.

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Figure 2. Ratio of elastic moduli as a function of the relative size of the medium.



Figure 4. Elastic modulus of the DNA-crosslinked gel as a function of crosslink density.