Measuring the Modulus of a Sphere by Squeezing between Parallel Plates

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Synopsis

Formulas are derived relating the deformation resulting from the application of a force f to the poles of a sphere of uniform modulus. When the force f is applied, the poles, originally a distance $2R^*$ apart, now are a distance $2R_v$ apart. Also, the diameter of the sphere at its equator is increased from $2R^*$ to a new value $2R_e$. For an ideal rubbery material with a shear modulus of G, the force is related to the change in equatorial diameter by

$$\sigma_c = \frac{f}{\pi R^{*2}} = G\left\{\left(\frac{R}{R^*}\right)_e^4 - \left(\frac{R^*}{R}\right)_e^2\right\}.$$

Thus, measurement of the increase in relative radius at the equator as a function of force applied at the poles yields the modulus directly. The change in R_v also can be related to G, but a numerical integration is required. Equations can also be derived relating modulus to dimensional changes when Hooke's law is used in place of ideal rubber elasticity.

INTRODUCTION

The modulus of a material can be measured using a variety of physical arrangements.[1,2] According to the classical model of an ideal rubber based on statistical mechanics, the shear modulus G, is identified as the product $N\boldsymbol{R}T$ where N is the moles of chains per unit volume, T is the absolute temperature, and $\boldsymbol{R}$ is the gas constant in appropriate units.[2,3]

For an ideal rubber in uniaxial tension or compression, the stress varies with change in length in a nonlinear fashion. For the case of compression, assuming that the volume does not change on stressing, one obtains:

$$\sigma_c = f/Ao = G(\alpha^{-2} - \alpha) \tag{1}$$

where the stress σ_c is the compressional force per unit unstrained, original cross-sectional area, and α the ratio of stressed to unstressed length, is less than one. On the other hand, for the case of simple shear, the shear stress is linear in the shear strain. Given the freedom to choose any geometry, shearing often is preferred since the response is linear and the shearing motion can be incorporated into a torsion pendulum or some other convenient apparatus.

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CHANGE IN EQUATORIAL DIAMETER OF A SPHERE ON COMPRESSION

Freedom of geometry is not always available. Consider the case of spherical samples, for example. To test the material in compression as a cylinder or in shear necessarily involves sacrificing the sample. The sphere has to be sliced up to obtain a cylinder or a slab with parallel surfaces. Uniform sections are difficult to produce. The very act of cutting or grinding may alter the physical properties. Most importantly, the sample is destroyed. Also, when the spheres are very small, cutting becomes impractical. If the sphere is compressed between two parallel plates and deformation is monitored as a function of force, the sample remains intact. An equation can be derived relating the force-deformation behavior to G.

Consider a sphere made of a material for which eq. (1) applies. First, consider a disc at the center (equator) of thickness $(dy^*)_e$ and radius R^* (Fig. 1). When the sphere is subjected to a compressive force f at the poles, the same force is transmitted through every layer of the sphere parallel to the equator. The thickness of the disc responds by changing from $(dy^*)_e$ to $(dy)_e$ in accordance with eq. (1)

$$(dy)_e = \alpha_e (dy^*)_e \tag{2}$$

where α_e is the strain ratio at the equator. We define a compressive stress σ_c at the equator based on the original undeformed area:

$$\sigma_c = f/(\pi R^{*2}) \tag{3}$$

The stress at the equator does not change when the sample is deformed since it is based on the original area. However, because the volume of the disc remains constant, the deformed disc has a new radius R_e, where

$$(R_e)^2 (dy)_e = R^{*2} (dy^*)_e \tag{4}$$

And, from eq. (2),

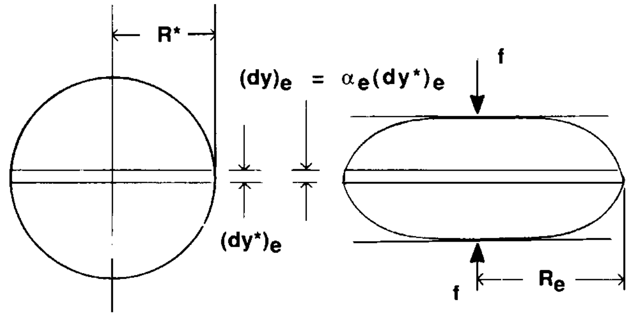

Fig. 1. The compressive force f deforms a disc of material at the equator decreasing the thickness $(dy^*)_e$ by a factor α_e and increasing the radius R^* by a factor of $R_e/R^* = (\alpha_e)^{-1/2}$ as the result of the total volume remaining constant.

$$(R^*/R)_e^2 = \alpha_e \tag{5}$$

Combining eq. (5) with eqs. (1) and (3) gives the relationship between force and change in equatorial radius (or diameter):

$$\sigma_c = \frac{f}{\pi R^{*2}} = G\left\{\left(\frac{R}{R^*}\right)_e^4 - \left(\frac{R^*}{R}\right)_e^2\right\} \tag{6}$$

Measurement of the relative increase in radius $(R/R^*)_e$ at the equator as a function of f yields the modulus G directly.

CHANGE IN TRANSPOLAR DISTANCE OF A SPHERE ON COMPRESSION

When a sphere is compressed, the distance between the poles changes much more than the equatorial diameter. This makes it a more sensitive measure of strain. A set of equations can be derived relating the force to the change in transpolar distance. Consider a disc of material with thickness dy^* at a vertical distance y^* from the center of the sphere (Fig. 2). The radius of the disc, x^*, may be obtained from:

$$x^{*2} + y^{*2} = R^{*2} \tag{7}$$

The compressive stress at y^* is

$$(\sigma)_{y^*} = f/(\pi x^{*2}) \tag{8}$$

and the local strain at y^* is

$$(dy) = \alpha(dy^*) \tag{9}$$

The vertical distance from the center of the sphere to the north pole R_v is

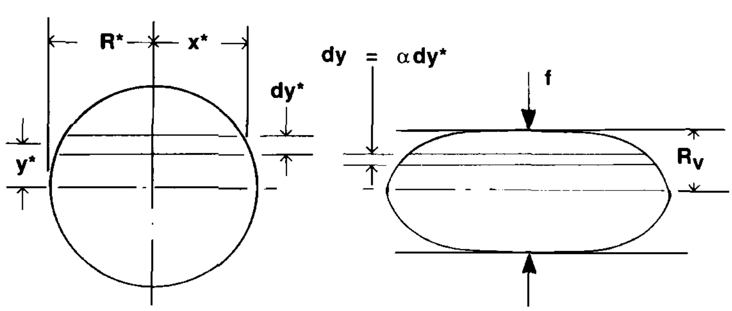

Fig. 2. The compressive force f deforms a disc of material a distance y^* away from the equator. The sphere is deformed into an oval cross-section with a transpolar distance of $2R_v$. β is defined as R_v/R^*.

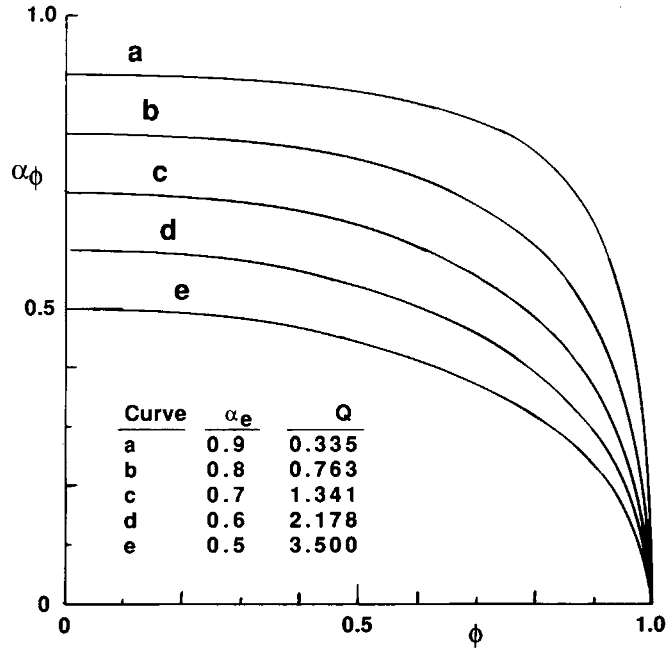

Fig. 3. The local vertical strain is a function of the original vertical displacement from the center of the sphere. ϕ = 0 corresponds to the center of the sphere, and ϕ = 1 corresponds to the polar cap. Q is the dimensionless stress at the equator.

$$R_v = \int_0^{v_{\max}} dy = \int_0^{R^*} \alpha \, dy^* \tag{10}$$

Because of spherical symmetry, the bottom half of the sphere (down to the south pole) is deformed identically, so that we need concern ourselves only with the top half of the sphere. The ratio of deformed diameter to original diameter is, of course, the same as the ratio of deformed radius to original radius. Both the change in total transpolar distance and the vertical position measured from the center of the sphere can be put in nondimensional form by normalizing with respect to the original radius:

$$\beta = R_v/R^* \qquad \text{and} \qquad \phi = y^*/R^* \tag{11}$$

Then eq. (10) becomes simply

$$\beta = \int_0^1 \alpha_\phi \, d\phi \tag{12}$$

Furthermore, using the equatorial stress σ_c from eq. (2), let

$$Q = \sigma_c/G \tag{13}$$

We can rewrite eq. (1) in terms of the equatorial compressive stress as

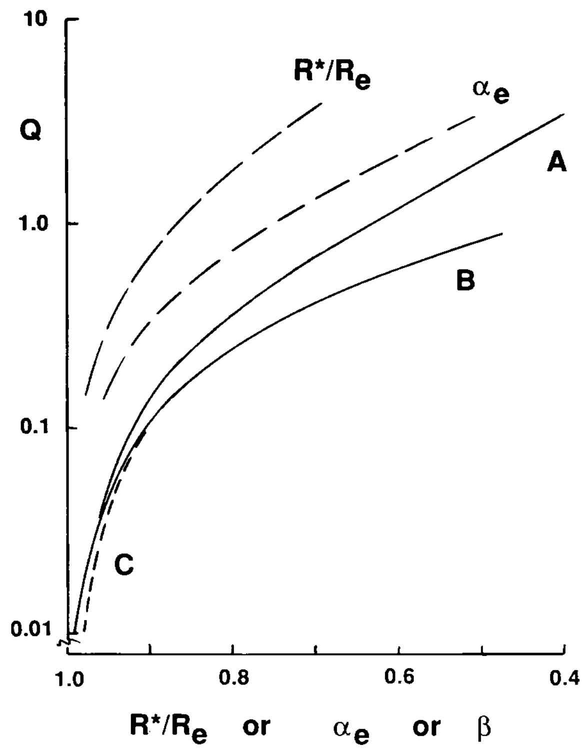

Fig. 4. Increasing dimensionless stress Q compresses the sample as seen by its effect on the dimensionless transpolar distance $\beta = R_v/R^*$. (A) The rubber elasticity model, eqs. (13) and (18); (B) Hooke's law, eq. (24); (C) Coalescence model, eq. (25), calculated only for the interval $0.9 < \beta < 1.0$. Also shown for the rubber elasticity model are the response of equatorial deformation R^*/R_e and its square, the equatorial strain, α_e.

$$Q = (\alpha^{-2} - \alpha)_e \tag{14}$$

At any latitude corresponding orginally to y^* (or ϕ), the *local* stress and *local* strain are related by:

$$(\sigma)_\phi = f/(\pi x^{*2}) = G(\alpha^{-2} - \alpha)_\phi \tag{15}$$

Also:

$$\frac{f}{\pi x^{*2}} = \frac{f}{\pi(R^{*2} - y^{*2})} = \frac{f}{\pi R^{*2}(1 - \phi^2)} = \frac{\sigma_c}{(1 - \phi^2)}\,. \tag{16}$$

Combining eq. (16) with (14) and (15) yields:

$$Q = (1 - \phi^2)(\alpha^{-2} - \alpha)_\phi = (\alpha^{-2} - \alpha)_e. \tag{17}$$

Thus, for each value of Q (the normalized equatorial stress), the local deformation, α_ϕ, at ϕ is a unique function of ϕ (Fig. 3). Local strain is always

TABLE I

Calculated Values from Numerical Integration of Rubber Elasticity Model

Equatorial radial strain, α_e, $(R/R^*)_e^2$	Transpolar radial ratio, $\beta = (R_v/R^*)$	Normalized equatorial stress, Q [eq. (15)]
0.50	0.3982	3.500
0.60	0.4858	2.178
0.70	0.5803	1.341
0.80	0.6858	0.763
0.90	0.8109	0.335
0.95	0.8882	0.158
0.96	0.9055	0.125
0.97	0.9244	0.0928
0.98	0.9450	0.0612
0.99	0.9677	0.0303
1.00	1.0000	0

greatest (α is smallest) at the polar surface ($\phi = 1$) for any value of Q greater than zero.

Numerical integration according to eq. (12) (the area under each α_e curve of Fig. 3) yields β which can be displayed as function of Q (Fig. 4, Table I). In the same figure, Q is shown as a function of the equatorial radius change, $(R/R^*)_e$ and of the square of that change, the equatorial strain, α_e. It can be seen that when the sphere has been compressed to half its original diameter, $\beta = 0.5$, the strain at the equator is only 0.62, corresponding to an increase in the diameter of only 1.27.

The shape of the deformed sphere can be calculated from the local values of x and y. The value of y can be obtained, by analogy to eq. (12), from the relationship

$$y/R^* = \int_0^{y^*} \alpha_\phi \, d\phi. \tag{18}$$

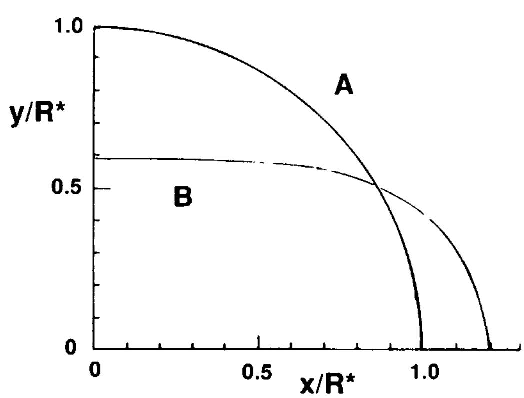

Fig. 5. Shape of a sphere, A, before, and B, after deforming to a Q value of 1.341.

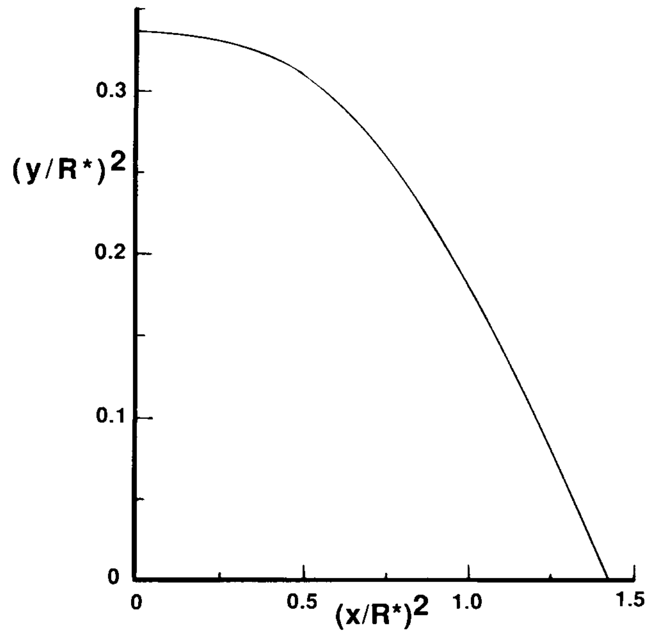

Fig. 6. Coordinates of deformed oblate spheroid surface plotted as squares. An ellipse would yield a straight line.

For x, we can use eq. (16) to express the local strain, α_ϕ, or $(x^*/x)^2$ as

$$\alpha_\phi = (x^*/x)^2(R^*/R^*)^2 = (R^*/x)^2(x^*/R^*)^2 = (R^*/x)^2(1-\phi^2) \quad (19)$$

Elimination of $(1-\phi^2)$ between eqs. (19) and (17) gives

$$(x/R^*)^2 = Q\alpha_\phi/(1-\alpha_\phi^3) \quad (20)$$

For the case where $Q = 1.341$ (curve c in Fig. 3), the x, y profile has been calculated (Fig. 5). That this is not an ellipse is most clearly seen by a plot of y^2 vs. x^2 which would be a straight line for an ellipse (Fig. 6). The oblate spheroid is observed in experiments with rubbery spheres.

COMPRESSION OF A LARGE SPHERE

A toy "superball" ("Teeny Bouncer," Imperial Toy Corp., Los Angeles, CA) was compressed using an Instron Testing Machine. The polymer in the ball presumably is a cross-linked polybutadiene. A compression load cell was mounted at the base of the machine and equipped with a flat plate. A tiny pin in the center of the plate kept the ball from slipping away. The cross-arm of the machine, also fitted with a plate, descended upon the ball at a speed of 0.50-in. (1.22 cm)/min. The original diameter of the ball was 1.00-in. (2.54 cm). Several balls gave very similar results. The compression was carried out to $\beta = 0.50$ and reversed several times. At the maximum compression the equa-

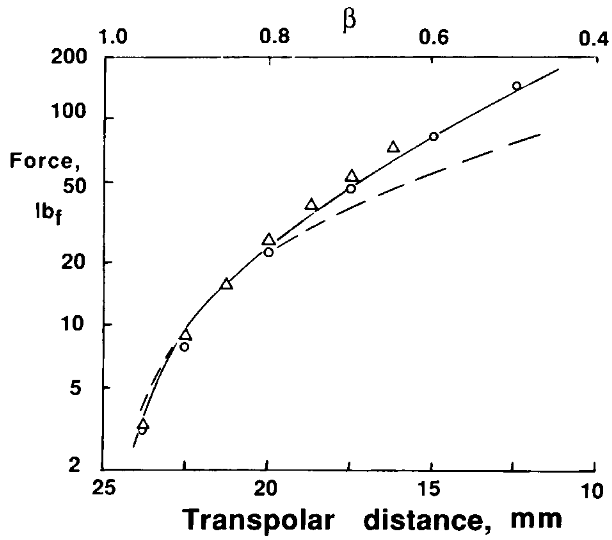

Fig. 7. Compression of "super-ball" in an Instron machine requires stout force. The circles represent the compression of a dry ball, the triangles are for a toluene-swollen ball. The solid line represents the rubber elasticity model fitted to the dry ball data. The dashed line is the fit by the Hooke's law model.

torial diameter had increased by a factor of 1.24 ± 0.03. There was no discernible hysteresis. Plotted as logarithm of force versus distance (Fig. 7), the experimental data are fitted by the theoretical Q,β line quite well. The force f_1 required to achieve $\beta = 0.64$ (where $Q = 1.00$) was used to calculate a modulus of 590 kPa (85 psi) from

$$G = f_1/\pi(R^*)^2 \tag{21}$$

One of the balls was equilibrated to a constant swollen volume over a period of two weeks in toluene at 23°C. The swollen diameter was 1.625 times the original. The volume fraction of polymer in the swollen state, v_2 was 0.233. At values of β greater than 0.65, the swollen ball could be squeezed with no discernible hysteresis (also shown in Fig. 7). However, at $\beta = 0.63$, the ball exploded into many fragments. Using the formulas given by Flory,[4] the tensile force f required to elongate an ideal rubber is given by

$$f/A_0 = (N\boldsymbol{R}T)_0(v_2)^{-1/3}(\alpha - \alpha^{-2}) \tag{22}$$

where A_0 is the unswollen cross-sectional area, N is the moles of polymer chains per unit volume in the unswollen sample, $\boldsymbol{R}$ and T are the gas constant and absolute temperature, and α is the ratio of stretched to unstretched length in the swollen state for a sample having a volume fraction of polymer in the swollen gel equal to v_2. The formula applies to dry or swollen rubber. Thus, if a dry sample ($v_2 = 1$) is elongated from a length of 10 cm to a length of 15 cm, then $\alpha = 1.5$, and the force required is

$$f_d = A_0(NRT)_0\,1.056 \tag{23}$$

If the same sample is placed in a solvent and swells isotropically to 8 times its original volume, v_2 is 0.125 and the new unstressed length is 20 cm. When the swollen sample is stretched to a length of 30 cm, α is 1.5 (based on swollen dimensions) and the force f_s should be

$$f_s = A_0(NRT)_0\,1.056(v_2)^{-1/3} \tag{24}$$

Thus $f_s/f_d = (v_2)^{-1/3} = 2.0$.

When the ball which was swollen in toluene was deformed, the force required was only slightly greater than that for the unswollen ball at the same fractional deformation. The prediction of eq. (22) is that the swollen sample should have required a force 1.625 times that for the unswollen sample. The reason for the discrepancy may lie in the fact that the chains in the superball are not usually in a random state when crosslinking occurs, since the pressure is often quite high. Therefore, the swelling in toluene may not be a straightforward test of eq. (22).

COMPRESSION OF SMALL GEL SPHERES

Cross-linked polyacrylamide gels were made by injecting droplets of aqueous monomer solutions into a continuous organic phase.[5] Individual gel spheres were compressed between Teflon® plates. The force was applied from the arm of an analytical balance and the deformation observed using a microscope fitted with an eyepiece micrometer. A representative result (Fig. 8) yields a modulus of 4.2 kPa. It is gratifying to see that the equation based on ideal rubber elasticity is as equally applicable to microscopic spheres as it is to macroscopic balls.

OTHER MATHEMATICAL MODELS

A much simpler mechanical model than eq. (1) is Hooke's law (with constant volume) which can be expressed as:

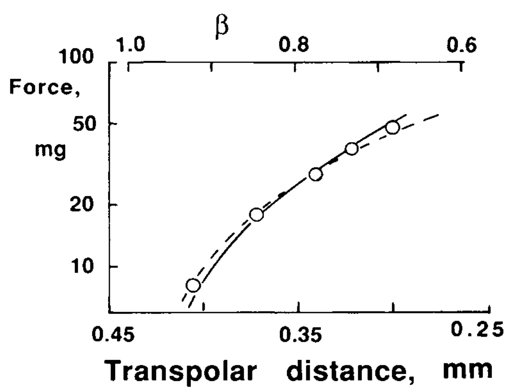

Fig. 8. Compression of a microscopic polyacrylamide gel particle. Masses in the milligram range are used to compress the particle which had an undeformed diameter of 0.441 mm. The solid line is the rubber elasticity model. The dashed line is the Hooke's law model.

$$\sigma_c = 3G(1 - \alpha) \tag{25}$$

where $G = E/3$ and E is Young's modulus in compression. At the equator, α_e again is simply $(R^*/R)_e^2$. Equation (17) is replaced by an equivalent expression through the same sequence of substitutions so that:

$$Q = 3(1 - \phi^2)(1 - \alpha)_\phi = 3(1 - \alpha)_e \tag{26}$$

An analytical solution is possible for β in this case. A complication in the integration step is that the compression ratio α has a lower limit of zero and cannot become negative. This means that the integration for β proceeds to $\phi = \phi_m$ rather than 1 where

$$\phi_m^2 = 1 - (Q/3) \tag{27}$$

Then integration according to eq. (12) yields:

$$\beta = \phi_m - (Q/6)\ln[(1 + \phi_m)/(1 - \phi_m)] \tag{28}$$

The Q,β plot for the Hooke's law case is illustrated in Figure 4. The experimental data are not very compatible with eq. (28). Of course, at very low deformations (β between 0.8 and 1), the simplicity of Hooke's law makes it easy to use and quite satisfactory from the standpoint of goodness of fit. The modulus values used to fit Hooke's law to the data of Figures 7 and 8 are about 1.5 times the rubber elasticity values (Table II).

Another approach has been used by workers who model the coalescence phenomena of latex particles under the influence of surface tension.[6] The end result is confined to values of β between 0.9 and 1 by the assumptions of the derivation. One such equation[6] can be put in the form of:

$$Q = K(1 - \beta)(1 - \beta^2)^{1/2} \tag{29}$$

With $K = 2.5$, the result is very much like Hooke's law (Fig. 7).

Finally, although restricted to small values of strain, the analysis of stress distribution in an epoxy sphere can shed some light on the actual variation within a sphere when it is subjected to uniaxial compression.[7] The material used, however, must be in the glassy state so that standard photoelastic techniques can be used at small strains to estimate the stress on "frozen" sections cut from the sphere.

TABLE II
Shear Moduli Derived from Experimental Data

Material	G (Hooke's law)	G (Elasticity theory)
Rubber sphere (Fig. 7)	840.0 kPa	590.0 kPa
Gel particle (Fig. 8)	6.3 kPa	4.2 kPa

CONCLUSIONS

The measurement of modulus by squeezing a sphere between parallel plates is a straightforward experimental approach which is applicable to rubbery samples of any size. While the equations derived on the basis of ideal rubber elasticity are quite satisfactory, they are restricted to materials which are crosslinked and amorphous. In general, the transpolar distance will be the more sensitive dimension to characterize deformation since it changes more than the equatorial diameter. The use of Hooke's law in place of ideal rubber elasticity is adequate when spheres are compressed only to about 0.8 the original transpolar distance.

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