SAND93-2058C
Conf. 940206-2

Measurement of Bulk Longitudinal Modulus and Correction for Edge Effects⁺

Daniel J Segalman
Structural Dynamics and Vibration Controls Department
and
Scott E. Klenke
Experimental Structural Dynamics Department

Sandia National Laboratories
Albuquerque, NM 87185

472504
0371

1.0 ABSTRACT

The measurement of the bulk longitudinal modulus, M^* , of rubbery polymers is a notoriously difficult task. Measurements are traditionally done through wave transmission through a fluid environment in methods that provide M^* over only very narrow frequency ranges. An alternative approach, using a resonant test fixture is presented in this paper. A discussion of some physical limitations is developed. Among them are issues of repeatability and edge effects. A method of data analysis to address edge effects is also presented.

2.0 INTRODUCTION

The complex bulk longitudinal modulus is the extensional modulus relevant to the calculation of acoustic impedance. It is the extensional modulus associated with oscillatory deformation in a longitudinal direction while lateral strain is precluded. This is the strain/loading configuration that one expects in a large, thin sheet attached on one side to a rigid surface and subjected to plane pressure waves on the other side. Representing the extensional stress as

$$\sigma(t) = \text{Im}(\sigma_0 e^{i\omega t}) \quad (1)$$

and the longitudinal strain as

$$\epsilon(t) = \text{Im}(\epsilon_0 e^{i\omega t}) \quad (2)$$

the complex bulk longitudinal modulus relates the two as

$$\sigma_0 = M^* \epsilon_0 \quad (3)$$

Since it defines the response of the material to a strain that involves a bulk deformation, the complex bulk-longitudinal modulus is very large for material of large bulk modulus. In fact, the complex bulk-longitudinal modulus M^* can be expressed in terms of the complex bulk modulus and the complex shear modulus: $M^* = K^* + \frac{4}{3}G^*$, where K^* is the complex bulk modulus and G^* is the complex shear modulus. Because the materials of interest in this investigation are rubbery polymers and such materials are nearly incompressible, the magnitudes of both K^* and M^* are very large. The above material properties are "complex" because they contain both magnitude and phase and are represented as complex numbers.

Various methods for measuring complex bulk longitudinal modulus are presented in Ferry¹. Additional acoustic methods involving "impedance tubes" are described by Capps². All these methods have their own limitations: restriction to very high frequency, requirement of extraordinarily high experimental accuracy, or the requirement of extraordinarily expensive and specialized

⁺This work, performed at Sandia National Laboratories, was supported by the U.S. Department of Energy under contract number DE-AC04-94AL85000

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

Segalman

equipment. It was the purpose of the work reported here to develop a method of routinely measuring complex bulk-longitudinal modulus with a table top device. That device and technique are presented below.

3.0 SANDIA DEVICE

The device, shown in Figure 1, consists of an electromagnetic shaker driving a plate on which rests a thin layer of the polymer and a reaction mass sitting above the polymer sample. Accelerometers are placed on both the plate and the reaction mass. The experimental configuration is designed to achieve strain only in the vertical direction, with no shearing strain or lateral displacement taking place. This configuration was described earlier by Rogers and Segalman³. The polymer sample has a thickness $2H$ and a diameter resulting in a cross-sectional area A .

FIGURE 1. Configuration of the device for measuring the transmissibility of the polymer layer, from which the complex bulk-longitudinal modulus can be calculated.

For rubbery polymers such as the materials which particularly interest us, the magnitude of K^* is much larger than the magnitude of G^* . For nearly incompressible materials, there will be a tendency for the polymer to shear toward the sides rather than to compress longitudinally and the relative motion of the plate and the reaction mass will result in erroneously small values for the bulk-longitudinal modulus. When testing such materials, it is important that the sample thickness be very small relative to the lateral dimension. Such a geometry exploits that the resistance to horizontal flow is on the order of $1/H^3$ whereas the resistance to vertical deformation is on the order of $1/H$.

The material response of the polymer is inferred by the magnitude and phase relations between the accelerometers in the manner derived below.

Referring to Figure 2, where the displacement of the base is represented as

$$u_1(s) = \text{Im} \left(e^{i\omega s} U_1 \right) \quad (4)$$

and the displacement of the reaction mass is represented as

$$u_2(s) = \text{Im} \left(e^{i\omega s} U_2 \right). \quad (5)$$

FIGURE 2. Kinematic quantities used in calculating the transmissibility of the polymer layer.

The kinematics of the experiment will be described entirely through the complex numbers U_1 and U_2 . Considering a free-body analysis of the reaction mass, the acceleration is expressed in terms of the applied loads:

$$-\omega^2 m U_2 = -A \sigma_0 \quad (6)$$

where, as before, σ_0 is the complex stress. Using the strain-displacement relationship

$$\epsilon_0 = (U_2 - U_1) / (2H) \quad (7)$$

and the stress-strain relationship involving $M^*(\omega)$ (Equation 3), the following expression is achieved for complex bulk longitudinal modulus in terms of the accelerometer responses U_1 and U_2 :

$$-\omega^2 m U_2 = -M^*(\omega) \left(\frac{A}{2H} \right) (U_2 - U_1) . \quad (8)$$

From the above, $M^*(\omega)$ can be expressed in terms of

$$e = U_2 / U_1 : \quad (9)$$

$$M^*(\omega) = \frac{2Hm\omega^2}{A(1 - 1/e)} . \quad (10)$$

Given the complex transmissibility, e , as calculated as the complex ratio of the two accelerations collected from the testing apparatus, the bulk longitudinal modulus is readily determined.

4.0 DEVICE LIMITATIONS

The outstanding limitations of the device described above are the numerical conditioning of Equation 10 and the validity of the postulated kinematics.

Equation 10 becomes ill-conditioned when e takes on either values near zero or near one. Those are cases where the upper mass remains nearly stationary while the lower mass oscillates and where the two masses move together, respectively. These two conditions are avoided when the excitation frequency is restricted to the vicinity of resonant frequency of the system composed of the polymer and the upper mass. This, however, is not a fundamental limitation since mass may be added or removed from the upper plate to investigate different frequency bands of interest.

There are four manners in which the postulated kinematics can be violated:

1. The excitation frequency can be so high that the gap occupied by the polymer is no longer small compared to the wave length of sound in the polymer. This is not a major concern since there exist other techniques that are well suited to measure complex bulk longitudinal modulus at high frequency¹.
2. The fixtures used to laterally restrain the moving element or armature can be excited creating rocking or pitching modes, producing some amount of lateral motion. When the test bandwidth coincides with frequency bands where lateral modes exist, it is more difficult to control the test. A further constraint of the experimental method is that it assumes that the excitation of the reaction mass remains in a linear range: stays in contact with the sample. These concerns are addressed by restricting the frequencies and amplitudes of excitation.
3. The large pressures developed within the polymer result in shear flow toward the free edges, creating a corresponding bulge and a corresponding increment in vertical displacement. Such extra vertical displacement will result in falaciously low estimates for the bulk longitudinal modulus. Also, the large shear deformation associated with this secondary flow would result in energy dissipation associated with the complex shear modulus. Since it is generally believed that the loss moduli in shear are much larger than those in bulk deformation, one expects the apparent phase of the "measured" bulk longitudinal modulus to be

FIGURE 3. Shear flow toward the free edges resulting in bulging.

larger than the true value for the material. A method of comparing the results of measurements taken with different sample geometries in order to resolve the above "bulging" effect is presented later.

4. Because the amount of shear is proportional to distance from the center of the sample, the shear stress also increases as one moves toward the free surfaces. If this results in "slip" between the polymer and the plates, the magnitude of the apparent bulk longitudinal modulus will be lower than that of the real material property. Similarly, one would expect that the dissipation associated with slip would also cause an overestimate of the phase angle. This particular violation of the postulated kinematics cannot be accommodated with refined data analysis. Instead, extreme care must be taken in sample preparation to preclude the likelihood of "slip" and equal care must be taken in data analysis to identify the cases for which slip has occurred none-the-less.

5.0 REFINEMENT OF DATA ANALYSIS

The analysis presented in this section was developed in order to facilitate the analysis of experimental M^* data in the presence of the shear deformation/ bulging kinematics discussed above. We consider a cylindrical coordinate system (r, θ, y) with its origin at the center of the polymer. The displacements with respect to the origin of the upper and lower plates are $v(t) = \text{Im}(v_0 e^{i\omega t})$ and $v(t) = -\text{Im}(v_0 e^{i\omega t})$, respectively. The displacement field in the polymer is postulated to vary linearly in the radial displacement and parabolically in the vertical direction:

FIGURE 4. Shear flow toward the free edges resulting in slip.

$$\underline{u} = \begin{bmatrix} g(r) \left[1 - \left(\frac{y}{H} \right)^2 \right] \\ 0 \\ v_0 \frac{y}{H} \end{bmatrix} \quad (11)$$

where $\underline{u}$ and $g(r)$ are complex. These kinematics are consistent with the "lubrication approximation" commonly used in hydrodynamics.

Ignoring inertial effects, as appropriate at low frequency, the momentum equation becomes

$$(1 - 2\nu) \nabla^2 \underline{u} + \nabla e = 0 \quad (12)$$

where

$$e = \nabla \bullet \underline{u} = \left(\frac{g}{r} + g' \right) \left[1 - \left(\frac{y}{H} \right)^2 \right] + \left(\frac{v_0}{H} \right) \quad (13)$$

and ν is the complex Poisson's ratio. Because the materials of interest are almost incompressible ν is almost equal to 1/2, and its actual value is very difficult to measure.

Substituting Equation 11 and Equation 13 into Equation 12, and then integrating through the thickness dimension results in only one non-trivial scalar equation:

$$(1 - 2\nu) \left\{ \left(\frac{1}{r} \right) \frac{d}{dr} [r g'] - 3 \frac{g}{H^2} \right\} + \frac{d}{dr} \left[\frac{1}{r} \frac{d}{dr} (r g) \right] = 0 \quad (14)$$

We also consider the stress boundary condition at the circumference of the polymer pad, $r = R$. This scalar equation is integrated over the thickness dimension to yield:

$$2\nu \left\{ \frac{4}{3} H \frac{1}{r} \frac{d}{dr} (r g) + 2 v_0 \right\} \bigg|_{r=R} + \left\{ (1 - 2\nu) \frac{4}{3} H g' \right\} \bigg|_{r=R} = 0 \quad (15)$$

Continuity requires that there be no radial displacement at the origin:

$$g(0) = 0 \quad (16)$$

The above system is now nondimensionalized:

$$r = \eta R, H = \frac{R}{\beta}, \text{ and } g(R\eta) = R \frac{v_0}{H} f(\eta) \quad (17)$$

We also introduce a parameter representing volumetric compliance:

$$\varepsilon = 1 - 2\nu = \frac{(G^*/M^*)}{1 - (G^*/M^*)} \quad (18)$$

The governing equation (Equation 14) becomes:

$$\frac{d}{d\eta} \left[\frac{1}{\eta} \frac{d}{d\eta} (\eta f) \right] + \varepsilon \frac{1}{\eta} \frac{d}{d\eta} (\eta f') - 3\varepsilon \beta^2 f = 0 \quad (19)$$

The boundary condition at the origin is:

$$f(0) = 0 \quad (20)$$

and the stress boundary condition at the circumference is:

$$(1 - \varepsilon)f(1) + f'(1) + (1 - \varepsilon)\frac{3}{2} = 0 \quad (21)$$

The net normal force is found by averaging the “y” component of stress through the thickness of the polymer and integrating over the area. A little manipulation shows the ratio of the measured bulk longitudinal modulus to the actual value to be

$$\frac{\hat{M}}{M^*} = 1 + \frac{4}{3} \frac{1 - \varepsilon}{1 + \varepsilon} \int_0^1 \left(\frac{f}{\eta} + f' \right) \eta d\eta \quad (22)$$

One observes that the dimensionless displacement function f is determined by the two dimensionless groups: ε and $\varepsilon\beta^2$. Though the compliance ε is no more known than M^* itself, a method will be presented below to exploit families of solutions to the above system of equations.

Solutions to Equation 19 can be found through the standard methods of series:

$$f(\eta) = \sum_{n=0}^{\infty} C_n \eta^n \quad (23)$$

Where the coefficients C_n satisfy the recurrence relation:

$$C_{m+2} = 3\varepsilon\beta^2 C_m / [(1 + \varepsilon)(m + 2)^2 - 1] \quad (24)$$

Because of the condition of Equation 20 $C_0 = 0$, and from the recurrence relation we see that all of the even subscripted coefficients are also zero. C_1 is scaled so that Equation 21 is satisfied

$$\sum_{n=1}^{\infty} C_n (1 + n - \varepsilon) + \frac{3}{2} (1 - \varepsilon) = 0. \quad (25)$$

The apparent bulk longitudinal modulus of Equation 22 is now related to the true modulus by:

$$\frac{\hat{M}}{M^*} = 1 + \frac{41-\epsilon}{31+\epsilon} \sum_{n=1}^{\infty} C_n. \quad (26)$$

A computer code to evaluate these series was written in the straight-forward manner. Numerical results plotting the relative error against $\epsilon\beta^2$ for several *real* values of ϵ are plotted in Figure 5. Also plotted there are several data points calculated by an elastic

FIGURE 5. Relative error of apparent M^* as a function of $\epsilon\beta^2$ for various real values of ϵ . The dots are values generated by a finite element computer code.

finite element code. One notices that the curves for the more nearly incompressible cases coalesce onto the curve generated by setting $\epsilon = 0$ wherever ϵ occurs alone in the above equations and is not multiplied by β^2 . In what follows, we refer to that case as the “incompressible limit”. One further observes very good agreement between the series solutions and the finite element solutions. Incidentally, the linear portion of the curve that occurs at high values of $\epsilon\beta^2$ is predicted by an analysis assuming that all shear motion is restricted to a boundary layer near the free surfaces.

Because the series solutions agree closely with those generated by a finite element code for the case of *real* ϵ , one has confidence in using the same series solutions for the complex case. Further, on the basis of the plots of Figure 5, we restrict ourselves to the case where ϵ is assumed to be zero wherever it is not multiplied by β^2 .

The effect of complex Poisson's ratio are explored in Figure 6, in which the magnitude of $\hat{M}/M^*$ is plotted as a function of $|\epsilon|\beta^2$ for various values of $\arg(G^*/M^*)$. These plots are all calculated at the “incompressible limit”, though ϵ is nonzero where it occurs in the product $\epsilon\beta^2$. It is seen that, at least as far as magnitude is concerned, the effect of the shear kinematics on the apparent bulk longitudinal modulus is independent of $\arg(G^*/M^*)$. The next figure, Figure 7, shows that the same is not true of the phase difference between $\hat{M}$ and M^* . It is shown in a following section how the insensitivity of the magnitude of $\hat{M}/M^*$ on $\arg(G^*/M^*)$

FIGURE 6. Magnitude of $\hat{M}/M^*$ as a function of $|\epsilon|\beta^2$ for various values of $\arg(G^*/M^*)$.

can be exploited to deduce the true value of $|\hat{M}/M^*|$ and $|\epsilon|\beta^2$ from experimental data. Experimental phase data is used with that value of $|\epsilon|\beta^2$ and Figure 7 to deduce the true $\arg(M^*)$.

6.0 REFINEMENT OF EXPERIMENTAL PROCEEDURE

One of the initial goals of the experimental method was to develop a process for testing the viscoelastic polymer material that had a high degree of repeatability. Originally, there were some repeatability problems related to the adhesion of the polymer material to the test fixture and reaction mass. The first polymer material experimentally tested was a polyurethane elastomer. A consistent and uniformly thick polyurethane sample could not be maintained when the adhesion process consisted of raising the temperature of the test apparatus until the polymer material was more plastic and soft enough to bond to the fixture and reaction mass. Further adhesion techniques used in conjunction with the polyurethane elastomer were one and two part epoxy bonds. The failure of the epoxy to produce a uniform bond across the contact surface between the polymer, test fixture and reaction mass made it difficult to attain repeatability. Due to the simplified test apparatus being used, this adhesion technique would also consistently create problems when the epoxy would flow around the edges of the polyurethane material before completely curing.

The importance of repeatability in the experimental method prompted the testing of spray and roll-on adhesives with a different polymer material, a polysiloxane elastomer. This method of adhering the polysiloxane elastomer to the test apparatus did show some promise, however, a homogeneous bond across the contact surface was still not achieved. Further investigation lead to the testing of a Scotch™ VHB™ (Very High Bond) Tape as a means of adhering the polysiloxane elastomer to the test apparatus. Although the polysiloxane material possessed a very smooth bonding surface, this family of adhesive transfer tape did provide a high level of repeatability in adhering this material to the test apparatus.

FIGURE 7. $\arg(\hat{M}/M^*)$ as a function of $|\epsilon|\beta^2$ for various values of $\arg(G^*/M^*)$.

By using a high tolerance mold in the production of the viscoelastic polymer material, the surface finish and thickness of the material can be reproduced with a high degree of reliability. In order to complete multiple experiments on similar viscoelastic material, two polymer test samples can be cut from each material lot produced. The test samples are cut with a hardened disk punch in order to produce a consistent material diameter. To ensure that thicker material lots do not bulge when they are cut, the temperature of the viscoelastic material is lowered, nearing the T_g (glass transition temperature) of the material, so that the material is less rubbery. Both the polymer test sample and the test apparatus then undergo surface cleaning before being bonded together with the Scotch™ VHB™ transfer tape. The ultimate bond strength of the tape is approached more rapidly by exposing the bond to an elevated temperature for two hours.

The data from the experiments is acquired using piezoelectric accelerometers. One accelerometer is located on the reaction mass and measures the acceleration response transmitted through the viscoelastic material. The other accelerometer, located on the base fixture, is used in the control of the experiment. The transfer function between the response on the reaction mass and the control is determined by the complex ratio of those two accelerometer signals. The complex transfer function is translated into a data file that can be read directly into MATLAB™ for further data analysis and reduction. A MATLAB™ m-file has been programed to calculate the bulk longitudinal modulus (as a function of frequency) using the transfer function data and material parameters such as area and thickness as input. A database of apparent bulk longitudinal modulus data has been generated for different materials, sample thicknesses, test levels and volumetric fill ratios. The experimental data is then integrated with analytic tools to compensate for the influences of edge effects.

7.0 A SAMPLE DATA SET

Three samples of a GE RTV 615 A+B were tested using the device described above. Samples thicknesses were 2.64 mm, 4.19 mm, and 8.18 mm, respectively, and all samples were 76.2 mm. in diameter. Each sample was tested twice, once with a 1.0 g acceleration and once with a 2.0 g acceleration. Figure 8, Figure 9, and Figure 10 show the measured bulk longitudinal modulus for the three

FIGURE 8. Measured bulk longitudinal modulus for the 2.64 mm thick sample of GE RTV 615 A+B.

samples over the frequency range 1200-1500 Hz. Only the plots associated with the 2.64 mm thick samples show variance between the 1.0 g and 2.0 g tests - a nonlinearity associated with slip between the sample and its sandwiching plates. By the same reasoning, one concludes that there is no slip in the cases of the thicker samples.

One observes that the apparent magnitude of the bulk longitudinal modulus decreases with increasing thickness. This is the relationship anticipated due to the shearing kinematics. The experimental data can be plotted against the experimental curves of Figure 6. The ordinate and abscissa are known up to unknown constant factors, since Figure 6 is log-log, one translates the three experimental data points vertically and horizontally to achieve best fit to the master curve. Focusing on that data at 1200 Hz, and giving no weight to the sample that appears to suffer slip, we find in Figure 11 that the data point associated with the thickest sample corresponds with an $|\epsilon|\beta^2$ value of $10^{-0.6}$. Since β is simply the aspect ratio, $|\epsilon|$ is computed to be 0.0029. More importantly, it shows that the asymptotic value of $|\hat{M}|$ is $10^{8.45}$ Pascals - ten times the value suggested by the raw data. Of course, there would be more confidence in the translations that put the experimental data on the master curve, if there were at least a third reliable data point.

Knowing the values of $|\epsilon|\beta^2$ corresponding to each data point, we can translate the experimental phase data vertically on Figure 7 until the points lie on one of the characteristic curves, indicating the true value of $\arg(G^*/M^*)$. This procedure is illustrated in Figure 12. We see that the first two data points lie in a range showing that $\arg(G^*/M^*)$ is approximately 0.35 radians. This, along with $|\epsilon|$ could be used to calculate the true $\arg(M^*)$, though less accumulation of error is likely if straightforward measurements of G^* are taken and $\arg(M^*)$ deduced from $\arg(G^*)$ and the above $\arg(G^*/M^*)$.

FIGURE 9. Measured bulk longitudinal modulus for the 4.19 mm thick sample of GE RTV 615 A+B.

FIGURE 10. Measured bulk longitudinal modulus for the 8.18 mm thick sample of GE RTV 615 A+B.

FIGURE 11. Magnitude of $\dot{M}/M^*$ as a function of $|\epsilon|\beta^2$ for various values of $\arg(G^*/M^*)$. Also plotted are the experimental data points taken at 1200 Hz.

FIGURE 12. $\arg(\dot{M}/M^*)$ as a function of $|\epsilon|\beta^2$ for various values of $\arg(G^*/M^*)$. Also plotted are the experimental data points taken at 1200 Hz.

8.0 CONCLUSIONS

The work presented here must be considered still a work in progress. Several difficulties inherent to the method have been identified and methods to address those difficulties have been developed.

Despite the difficulties, it appears clear that the experimental method presented here has the capability to provide measurements of bulk longitudinal modulus in a table-top environment.

9.0 REFERENCES

1. J. D. Ferry, *Viscoelastic Properties of Polymers*, John Wiley and Sons, 1980, pp 171-175.
2. R. N. Capps, *Elastomeric Materials for Acoustical Applications*, Naval Research Laboratory, September 1989, pp 33-37
3. J.D. Rogers and D. J. Segalman, "A Method for the Measurement of the Complex Compressional Modulus of thin Layers", presented at Damping 91, Feb 13-15, 1991, San Diego, CA.

DATE

FILMED

2 / 24 / 94

END