Cell poking:
Quantitative analysis of indentation of thick viscoelastic layers

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ABSTRACT A recently introduced device, the cell poker, measures the force required to indent the exposed surface of a cell adherent to a rigid substratum. The cell poker has provided phenomenological information about the viscoelastic properties of several different types of cells, about mechanical changes triggered by external stimuli, and about the role of the cytoskeleton in these mechanical functions. Except in special cases, however, it has not been possible to extract quantitative estimates of viscosity and elasticity moduli from cell poker measurements. This paper presents cell poker measurements of well characterized viscoelastic polymeric materials, polydimethylsiloxanes of different degrees of polymerization, in a simple shape, a flat, thick layer, which for our purposes can be treated as a half space. Analysis of the measurements in terms of a linear viscoelasticity theory yields viscosity values for three polymer samples in agreement with those determined by measurements on a macroscopic scale. Theoretical analysis further indicates that the measured limiting static elasticity of the layers may result from the tension generated at the interface between the polymer and water. This work demonstrates the possibility of obtaining quantitative viscoelastic material properties from cell poker measurements and represents the first step in extending these quantitative studies to more complicated structures including cells.

INTRODUCTION

Studies of the mechanical properties of cells are important for an understanding of their function in many physiological and pathological processes. This is obvious for muscle and bone cells, but it is also true for many other cell types. For example, blood cells must deform to pass through the microcirculation and to traverse capillary walls into peripheral tissues (Bagge and Branemark, 1977, Meiselman et al., 1984; Chien, 1987; Worthen et al., 1987). Other cells such as ova (Vacquier, 1981), secretory cells (Pfeiffer et al., 1985), or cells of the immune system respond dynamically to external signals by changing shape and cytoskeletal organization (Bourguignon and Bourguignon, 1984; Howard and Oresajo, 1985). The role of these mechanical changes in the overall physiological process is not yet clear. Measurements of cellular viscoelasticity can illuminate these events from a new perspective and can provide important functional information about the physiological role and mechanical-structural properties of the cytoskeleton (Elson, 1988).

Previous studies of cellular deformability have mainly emphasized micropipette aspiration measurements of blood cells. Methods for measuring red cell deformability have recently been evaluated (Staubli et al., 1986). Recently, a new method, called "cell poking," has been developed, which is well adapted to measuring deformability both of blood cells and of adherent cells (Petersen et al., 1982). A fine glass stylus is used to indent the exposed surface of a cell adherent to a rigid substratum. The force of the cellular resistance to indentation is registered as a function of the depth of indentation. This method has been applied to study erythrocytes (Daily et al., 1984), fibroblasts (Petersen et al., 1982; Daily, 1984; Cooper et al., 1987), lymphocytes (Pasternak and Elson, 1986; Schwab et al., 1987), RBL cells (Liu et al., 1987), and pancreatic islet cells (Schwab et al., 1986). In general it has been observed that both viscous and elastic forces resist deformation at the rates of stylus motion which are typically used.

Ideally cell poking data should be interpreted to yield quantitative measures of the viscosity and elasticity of the cell, that is, of the mechanical properties of the cell's constituent materials independent of cell geometry. The complexities of shape and mechanical characteristics of typical cells make this a difficult undertaking in general. Hence cell poking measurements obtained up to now have been analyzed mostly in terms of an empirical stiffness parameter (e.g., Pasternak and Elson 1985; Liu et al., 1987). To develop a more fundamental mechanical interpretation of the data we have begun to carry out measurements on mechanically well defined samples that are confined to relatively simple shapes. Here we present the results of a cell poker study of a relatively simple system, well characterized polydimethylsiloxane (PDMS) melts confined to planar layers which are thick compared with

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0006-3495/89/04/683/08 $2.00

the stylus diameter. The aims of the present study are as follows: first, to validate this experimental technique using a well defined viscoelastic material; second, to present the theory of the deformation of the viscoelastic half space by a rigid cylindrical indenter; and third, to begin to establish a theoretical basis for the interpretation of cellular deformability data using the deformation of a half space as a model. We shall show that microscopic cell poker measurements of the PDMS layers yield values of the mechanical properties which agree with independent determinations on a macroscopic scale of the relevant PDMS parameters, except that surface forces, which are presumably negligible in large scale measurements, appear to be important on the scale of the cell poker.

The next step here will be the extension of the theory and experimental measurements to include the deformation of thin layers and to spheres, which should provide approximate analogues to relatively flat and round cells such as fibroblasts and lymphocytes respectively, and then ultimately to bodies of arbitrary shape.

MATERIALS AND METHODS

The three linear PDMS used here, although manufactured by Dow Corning (Midland, MI), were obtained from two different sources. Samples designated A and C were obtained from Sigma Chemical Co., (St. Louis, MO), sample B was purchased from Accumetric (Elizabethtown, KY). Characteristics of these samples, obtained from the producer are summarized in Table 1.

The layers were prepared by pouring PDMS between two coverslips and sliding the coverslips apart. The thickness of the layers used was >400 μm. They were then allowed to stand at room temperature overnight and used the following day. The poker chamber was filled with a 0.2% bovine serum albumin (wt/wt) (Sigma Chemical Co.) in filtered, deionized water. The serum albumin decreased adhesion between the poker tip and the layer.

At the beginning of the experiment the layer was inverted on the chamber, and the layer and the medium in the poker chamber were allowed to reach thermal equilibrium (25 ± 10 min) at room temperature (~20°C).

The cell poker has been described previously (Petersen et al., 1982). Briefly, a vertical glass fiber, ~25 μm in diameter in these experiments, mounted at one end of a horizontal glass beam (~3 cm in length and 75 μm in diameter) was used to indent the layer. The beam used in these experiments had a stiffness of 0.039 N/m. The other end of the horizontal beam was attached to a linear piezoelectric motor that moved the beam up and down. The vertical position of the poker tip was monitored accurately by an optical sensor that measured the light reflected from a gold coated mylar flag mounted at the base of the tip. As the tip rose, the flag moved out of the light beam so that less light was reflected to the sensor. In the experiments described here a triangular voltage waveform was fed to the motor as a command signal and the position of the tip was monitored. The experiments were performed at several motor velocities (from 0.98 μm/s to 15.6 μm/s) and several amplitudes of motor displacement (8–32 μm).

TABLE 1 Characteristics of PDMS Samples‡

Sample	Viscosity	M_n*	M_w*	M_w/M_n
	Ns/m²			
A	12.5	22,000	56,000	2.5
B	30.0	24,000	78,000	3.2
C	60.0	28,000	97,000	3.5

*M_n and M_w are the number and weight average molecular weights, respectively.

‡This information was kindly supplied by Dr. Anthony P. Wright, Dow Chemical Corp., Midland, MI.

THEORY

Consider an elastic layer of thickness h lying on a rigid half space and firmly connected to it. Let the layer be deformed by an axisymmetric flat-ended rigid poker of radius a, as shown in the Fig. 1. The relation between applied force F and the depth of indentation u based on linear elasticity theory has been given by several authors (for reviews see Gladwell 1980, Johnson 1985), and can be expressed in the following form:

$$F = 4Gau\alpha(a, h)/(1 - \nu), \quad (1)$$

where G is the elastic shear modulus, ν is Poisson ratio, and α is a function of poker radius and the layer thickness. Surface tension effects are ignored in this calculation.

When $a \ll h$ and for an incompressible material ($\nu = 0.5$), the function α is well approximated by a series:

$$\alpha = 1 + 1.12696\epsilon + 1.27003\epsilon^2 + 1.18369\epsilon^3 + 1.05496\epsilon^4 + 0(\epsilon^5), \quad (2)$$

where $\epsilon = a/h$. This equation has been derived from the results obtained by Dhaliwal et al. (1977). In all our experiments, ϵ is <0.05 and so we assume that α is equal to 1.

The solution for the indentation of a viscoelastic layer can be obtained from the elastic solution by applying the correspondence principle (Bland, 1960; Christensen,

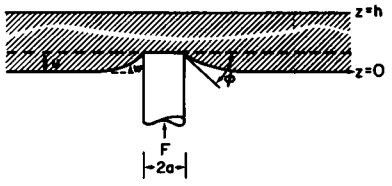

FIGURE 1 Deformation of the layer of thickness h, resting on a rigid base. G is the elastic shear modulus and ν is the Poisson ratio of the layer. As discussed in Appendix B, w, u, F, and ϕ represent the upward displacement of the deformed interface from its unloaded position, the depth of indentation, the indenting force, and the angle of the indented surface at the edge of the poker respectively.

1982). According to this principle, the Laplace transformed viscoelastic solution can be obtained directly from the elastic solution by replacing G and ν with $sG(s)$ and $(3\kappa[s] - 2G[s])/(2s(G[s] + 3\kappa[s]))$, respectively, where $\kappa(s)$ and $G(s)$ are the Laplace transforms of relaxation functions in dilatation $\kappa(t)$, and shear $G(t)$. It is possible to apply the correspondence principle in this case because the area of contact between the poker tip and PDMS layer is assumed to remain constant during the indentation.

If we assume that the viscoelastic layer is incompressible ($\nu = 0.5$) Eq. 1 yields:

$$F(t) = 8a \int_0^t G(t - \tau)\, du(\tau). \tag{3}$$

Because $u(t)$ and $F(t)$ are measured experimentally, Eq. 3 can be solved in order to find $G(t)$. The numerical procedure used to solve the above equation for $G(t)$, is given in Appendix A.

Any reasonable relaxation function can be written in the following general form:

$$G(t) = G_\infty + G_1(t), \tag{4}$$

where G_∞ is a non-negative constant (equal to 0 for viscoelastic fluids), and $G_1(t)$ is a decreasing function of time, and $\int_0^\infty G_1(t)\, dt = \eta$ ($\eta < \infty$). By analogy with linear viscoelastic liquids (Christensen, 1982), we can designate η as the effective viscosity of the PDMS layer. Assuming that the velocity of the stylus tip is constant (which is true for all but extremely short times in our experiments) and is equal to v, Eq. 3 becomes:

$$\begin{aligned} F(t) &= 8av \int_0^t [G_\infty + G_1(\tau)]\, d\tau \\ &= 8av \left(G_\infty t + \int_0^\infty G_1(t)\, dt - \int_t^\infty G_1(\tau)\, d\tau \right) \\ &= 8av \left(G_\infty t + \eta - \int_t^\infty G_1(\tau)\, d\tau \right). \end{aligned} \tag{5}$$

If the time t tends to infinity (i.e., $t \gg \tau_e$, where τ_e is the longest relaxation time of the sample), the indentation proceeds at constant velocity; the last term drops out; and Eq. 5 can be written in the following form:

$$F(t)/(8av) = G_\infty t + \eta. \tag{6}$$

The above equation shows that at $t \gg \tau_e$ force is a linear function of the time. The elasticity and viscosity parameters G_∞ and η can be determined directly from the slope and the $t = 0$ intercept respectively of the asymptotic linear region of a plot of $F(t)/(8av)$ versus time. Note that this characterization of the viscoelastic response to ramp indentation is independent of any particular assumption about the form of the relaxation function.

In all the experiments described here the shape of the driving function for the motor is given by a triangular wave form $u(t)$:

$$u(t) = u_0(1 - |t/T - 1|), \quad 0 \le t \le 2T, \tag{7}$$

where u_0 is the maximum amplitude of the indentation, and T is the half period. This means that the motor moves an unloaded poker tip uniformly to the depth u_0 (at time $t = T$), and then returns it to the starting position $u = 0$ (at time $t = 2T$). For linear, ideally elastic materials the plot of displacement versus force would be linear, with the force at any tip displacement being equal for both increasing and decreasing deformation. For viscoelastic bodies, viscous effects cause a dissipation of energy which is equal to the hysteresis area during one cycle. The total energy, D, dissipated during one cycle of deformation is

$$D = \int_0^{2T} F(t)\dot{u}\, dt. \tag{8}$$

Hence, this area of hysteresis can be regarded as an independent measure of viscous effects; it can be calculated using Eq. 8 for any assumed $G(t)$. The hysteresis area, however, depends very strongly on the shape of the driving function $u(t)$. Because the accuracy of the approximation given by Eq. 7 varies with velocity and depth of indentation, this calculation is not very accurate (especially for slow velocity and small indentation), and has been considered as a qualitative measure only.

As shown below, our measurements yield a finite value for the limiting elastic modulus G_∞ even though macroscopic samples of PDMS have been shown to behave as viscoelastic liquids Eq. 2. We attribute this elasticity to the interfacial tension generated at the water-PDMS interface which is present in our experiments. We suppose further that the interface influences only the elastic contribution, G_∞, and does not significantly affect η. An analysis of the contribution of the interfacial tension to G_∞, which corroborates these assumptions, is presented in Appendix B.

RESULTS

Typical results are presented in Fig. 2 *a* which shows five sets of data in which reference curves are paired with corresponding displacements of the poker, measured as it indented the layer. The reference curves represent the displacement of the stylus in response to the command signal in the absence of contact with the layer. Fig. 2 *a* shows excellent agreement at corresponding extents of indentation among measurements obtained from separate experiments. The difference between the paired reference and measured curves allows us to determine the bending

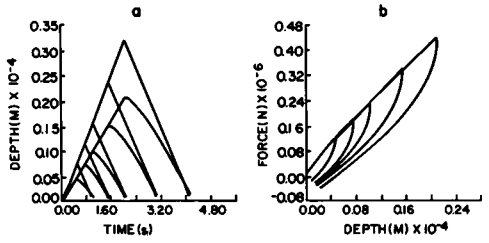

FIGURE 2 Cell poker measurements of layers of PDMS from sample C. The velocity of deformation is 15.6 μm/s. Panel A. Depth of indentation versus time. Five pairs of measurements are shown. In each pair the reference curve (to larger depth) shows the trajectory of the unloaded probe in response to the triangular command signal. The corresponding trajectory of the probe when in contact with the PDMS layer reaches a smaller depth due to the resistance of the layer. Panel B. The data shown in Panel A are replotted as to display force versus depth of indentation.

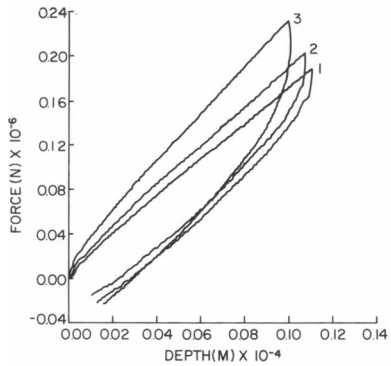

FIGURE 4 The effects of PDMS viscosity on force versus deformation curve. Curve 1: sample *A*, curve 2 sample *B*: curve 3: sample *C*. For all measurements the motor velocity = 15.6 μm/s, and the maximum motor displacement was 16 μm.

of the beam. Because the stiffness of the beam is known, computation of the force necessary to produce this bending is straightforward. The results of such calculations are shown in Fig. 2 *b* as a plot of force versus depth of indentation. It can be seen that hysteresis area increases with the depth of indentation.

The dependence of the force-indentation plots on the velocity of the poker tip is represented in Fig. 3. As expected from the dependence of viscous resistance on velocity, both the measured force decreases with decreasing indentation velocity and the maximum depth of indentation increases. The hysteresis area is approximately proportional to the velocity of deformation over the range investigated.

Fig. 4 shows the comparison of force versus indentation plots for different PDMS viscosities (samples A, B, and C). The influence of viscosity is similar to that of the velocity as expected for a linear system. The decrease in viscosity causes a decrease in the force exerted on the poker tip by the layer at a given indentation. Similarly, the hysteresis area increases with increasing viscosity. Figs. 4 and 5 show that the layer can exert negative force on the probe at the end of the withdrawal phase of the indentation cycle. We attribute this to adhesion of the poker tip to the layer surface. In evaluating the hysteresis area negative force values were set to zero. The existence of an adhesion force between the glass probe tip and the layer supports our assumption of nonslip contact between the glass surfaces and the layer.

The relationship between the velocity of deformation and force at a fixed time (t = 0.5 s) is given in Fig. 5. The data are fitted very well by a straight line through the origin in agreement with Eq. 6.

The elastic and viscous parameters of the layers have been determined by fitting all the experimental data to

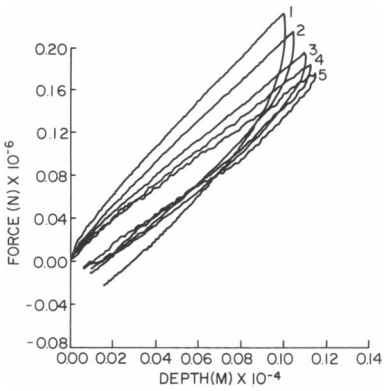

FIGURE 3 The effects of changing probe velocity on force versus deformation curves. The measurements were carried out on PDMS layers from sample C. The maximum displacement of the motor was 16 μm. The motor velocities used in the measurements were curve 1: 15.6 μm/s, curve 2: 7.8 μm/s, curve 3: 3.9 μm/s, curve 4: 1.95 μm/s, curve 5: 0.98 μm/s.

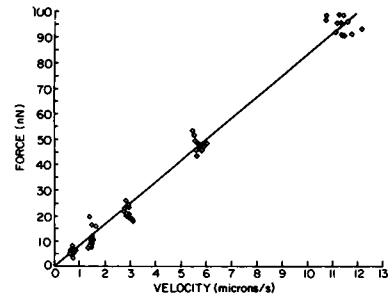

FIGURE 5 Force versus probe velocity. Measurements were carried out on PDMS layers from sample A. Each point shows the force at time = 0.5 s.

Eq. 6. The results are given in Table 2. It can be seen that G_∞ shows less measurement variability than η.

Using the method presented in Appendix A, it was possible to determine the relaxation function $G(t)$ from data like those shown in Fig. 2 *b*. The results of these calculations are plotted in double logarithmic scale in Fig. 6. We can distinguish two limiting constant values of $G(t)$, G_0 and G_∞, the instantaneous and long-time elastic moduli. The former exceeds the latter owing to viscous forces which resist indentation at high rates and which diminish as the rate of indentation diminishes. The length of the G_0 plateau increases with the viscosity of the polymer (or molecular weight), whereas the G_∞ plateau starts earlier the less viscous (the lower molecular weight) the polymer. The area under the curve of time versus $(G(t) - G_\infty)$ can be interpreted as the steady state viscosity η, (Ferry, 1980). Numerical values of G_∞, G_0 and η are given in Table 3. There are no significant differences between the G_∞ values or between the G_0 values for polymers A, B, and C. One should note that there is good agreement between the data given in Tables 3 and 2. Also the values of η presented in Tables 2 and 3 are in good agreement with the nominal values determined by standard methods (Table 1).

DISCUSSION

Our overall objective in this work was to validate the quantitative measurement by the cell poker of the viscoelastic properties of a well characterized substance having a simple, well defined shape. This also provides a starting point from which the analysis may be extended to more complicated substances and shapes which may more closely approximate cells. The theoretical analysis of cell poker measurements of thick viscoelastic layers has been confirmed experimentally. PDMS was chosen as a test material because it has been well characterized by other experimental studies (Barlow et al., 1964; Rahalker et al., 1984; Queslel and Mark, 1984). Also the viscosity of this material is of the order of that of cells (Chien et al., 1984).

The layers used in the experiments were thick enough to neglect the presence of the rigid substratum to which the layer was attached. The maximum depth of indentation of the layer was ~24 μm which is comparable with the poker diameter and much smaller than the layer thickness (>400 μm). Experiments in which the cell poker was driven by a sinusoidal command signal show that all measurements were within the range of linear response (unpublished data). Also the data presented in Fig. 5 show that the force has a linear dependence on the velocity of indentation. This result agrees with prediction from the theory of linear viscoelasticity (Eq. 6) and justifies our application of that theory to the analysis of these experiments. Nevertheless, it was somewhat surprising that the linear theory serves so well despite the fact that the indentation depths were not small compared with the poker diameter. As discussed below, the influence of the interfacial tension at the PDMS surface may account for the observed linearity to some degree.

The data presented in Figs. 3 and 4 show the influence of the velocity of indentation and the viscosity of the layer on the resistance to deformation. An increased velocity causes a decrease of the depth of displacement similar to that resulting from an increased PDMS viscosity.

The shape of the relaxation function presented in Fig. 6 agrees qualitatively with the predictions of the molecular theory of polymer viscoelasticity (Doi and Edwards, 1986). The width of the G_0 plateau increases with increasing molecular weight, M (see Tables 1 and 3), and the

TABLE 2 Viscosity and elasticity parameters of PDMS samples from Eq. 6

Sample	G_∞	η
	Pa	*Pa s*
A	136.1 ± 14.4	14.5 ± 7.5
B	133.6 ± 12.8	24.8 ± 13.8
C	148.1 ± 15.7	65.2 ± 21.1

TABLE 3 Viscosity and elasticity parameters of PDMS samples from Fig. 6

Sample	G_0	G_∞	η
	Pa	Pa	Pa s
A	416.6 ± 89.7	130.8 ± 12.3	15.4
B	431.4 ± 81.9	133.5 ± 8.3	34.7
C	438.5 ± 78.4	135.2 ± 8.6	78.4

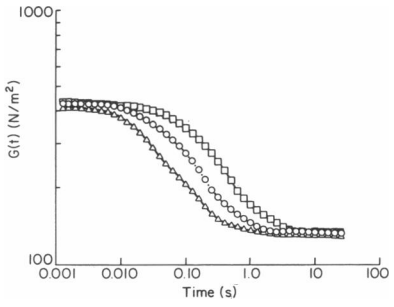

FIGURE 6 Plots of the relaxation functions versus time. △: sample A, ○: sample B, □: sample C. The relaxation functions were derived from the experimental data using the approach of Appendix A.

height of plateau is independent of M, in agreement with theoretical predictions, (Doi and Edwards, 1986). The nonzero value of elastic plateau modulus G_∞, indicates that the samples of PDMS behave as viscoelastic solids rather, than viscoelastic liquids. The measured G_∞ is most likely due to the interfacial tension at the PDMS-water interface. The observation that G_∞ is the same for the three PDMS samples is consistent with the expectation that the interfacial tension between water and a polymer melt should be independent of the molecular weight of the polymer (Wu, 1982). In Appendix B we present an analysis of the forces associated with the interfacial tension. Using measured values of the density and surface tension of commercially available PDMS and a poker radius of 12.5 μm, we have calculated that the contribution to G_∞ from this source is 278 Pa. This value is the minimum G_∞ that one would expect to measure, even if the G_∞ of the bulk PDMS were zero. In fact our measured values of G_∞ were lower, ~135 Pa. One possible explanation for this discrepancy is that additional solution components, especially the bovine serum albumin, which we added to decrease adhesion of the poker tip to the PDMS layer and which might have formed a surface film on the PDMS, lowered the interfacial tension to around one half the value of 30 dynes/cm used in Appendix B. Thus the fact that PDMS appears to behave as a viscoelastic solid in cell poker experiments, rather than as liquid is a consequence of interfacial tension, a factor which becomes negligible when the material is tested in bulk. Indeed when mechanical tests are performed on a cellular scale one must always expect that interface forces may play an important role, and it is gratifying that in the present experiments we have been able to identify and characterize these forces. In animal cells the plasma membrane will govern the mechanical properties of the boundary between the cell and the medium. This membrane is structurally complex with polar lipid head groups and embedded glycoproteins and glycolipids that will modulate interactions of the membrane with the medium and an underlying cytoskeletal cortex that can generate tension (Bray et al., 1986; Chasis and Shohet, 1987; Elson, 1988).

One of the possible methods for the analysis of the data would be to fit it to a simplified model such as the viscoelastic standard solid (Fung, 1965) as has been done for micropipette aspiration of cells (Schmid-Schonbein et al., 1981). When this was done, the fit to individual experiments was excellent as shown in Fig. 7. However, the coefficients of the model were depth and velocity dependent when this analysis was applied to the whole data set (unpublished data). This might result from a complex dependence of the viscosity and elasticity parameters of the model on molecular properties. For example representation of a Rouse-Mooney viscoelastic solid in terms of the linear standard model yields spring constants and a viscosity which depend on sums of exponential functions of the polymer relaxation times (Elson, 1988). The analyses based on Eq. 6 and on Appendix A, however, do not assume a mechanical model and require only very general assumptions about the shape of the relaxation function.

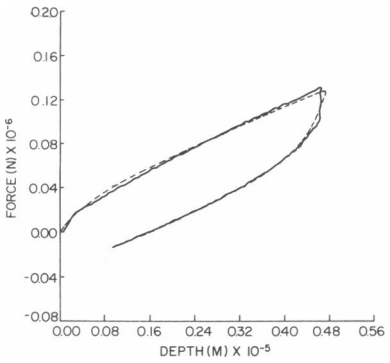

FIGURE 7 Fitting of viscoelastic standard model to experimental data. In this example the dashpot had a viscosity of 34.5 Pa s. The springs in parallel and series with the dashpot had spring constants of 198 Pa and 458 Pa. The continuous and dashed curves represent the experimental data and model fitting respectively.

CONCLUSIONS

The cell poker, using constant-velocity indentation ramps, yields reproducible and consistent measurements of the viscoelastic parameters of thick PDMS layers for a wide range of amplitudes (up to 32 μm), and velocities (up to 15.6 μm/s).

Even for deformations comparable with the poker diameter, the measured responses agreed with the linear viscoelasticity theory applied to analyze the deformation of the PDMS layers. The calculations in Appendix B help to explain this somewhat unexpected result.

Two methods for calculating the viscoelastic parameters from cell poker measurements have been presented. One is based on numerical calculation of the relaxation function from the experimental data. The other, convenient and relatively accurate, requires that the force response becomes steady, and therefore that the time of experiment be longer than the longest relaxation time of the material. Calculated values of viscosity by both methods were in good agreement with nominal values.

The relaxation function of PDMS was in qualitative agreement with the predictions from the molecular theory of viscoelastic solids. The measured static elasticity G_∞, can be explained by the presence of interfacial tension between the PDMS and water.

APPENDIX A

Numerical approximation of the relaxation function

Let us break up the integral in Eq. 3 into a finite number of integrals over small increments of time,

$$F(t) = 8a \sum_{i=1}^{n} \int_{t_{i-1}}^{t_i} G(t - \tau)\, dz(\tau).$$

If we assume that $G(t)$ is a smooth function, then the above equation can be approximated by

$$F(t) = 4a \sum_{i=1}^{n} [G(t - t_i) + G(t - t_{i-1})][(z(t_i) - z(t_{i-1})].$$

This equation can be solved for $G(t_n)$, yielding the following numerical approximation to the relaxation function,

$$G(t_n) = \frac{F(t_n) - 4aG(0)[z(t_n) - z(t_{n-1})] - 4a \cdot \sum_{i=1}^{n-1} G(t_i)(z(t_{n+1-i}) - z(t_{n-1-i}))}{4a[z(1) - z(0)]}.$$

APPENDIX B

Contribution of interfacial tension to G_∞

Consider the static indentation of a layer of PDMS by a flat-ended, rigid cylindrical poker of radius a, as shown in Fig. 1. The layer is bounded above by a flat rigid surface (the glass cover slip) and below by a reservoir of water. We assume that under static conditions the PDMS behaves as a liquid and can not support shear stresses. We assume further that the interface between the water and the PDMS behaves mechanically as a membrane with constant interfacial tension T. Let w represent the upward displacement of the deformed interface from its unloaded position, u the depth of indentation, and F the indenting force. Further let ρ_g and ρ_w represent, respectively the densities of PDMS and water, p_0 the pressure at the interface at points remote from the poker, and ϕ the angle of the membrane at the edge of the poker. In the plane $w = 0$ we can establish cartesian coordinates (x, y) or polar coordinates (r, θ).

The pressure increase across the interface (in passing from water to PDMS) is simply computed from hydrostatics as

$$\Delta p = (p_0 - g\rho_g w) - (p_0 - g\rho_w w) = gw\Delta\rho, \qquad \text{(B1)}$$

where g is the gravitational acceleration and $\Delta\rho = \rho_w - \rho_g$. The well known equation of membrane equilibrium (Novozhilov, 1959) requires that at each point of interface

$$\Delta p = T(R_1^{-1} + R_2^{-1}), \qquad \text{(B2)}$$

where R_1 and R_2 are the principal radii of curvature. Thus the quantity in parentheses is twice the mean curvature of the interface, which can be approximated as

$$\nabla^2 w = w_{,xx} + w_{,yy} = w_{,rr} + r^{-1}w_{,r} + r^{-2}\, w_{,\theta\theta} \qquad \text{(B3)}$$

if the normal to the interface is everywhere close to vertical. Adopting this last approximation, and assuming that the deformed interface is cylindrically symmetric (i.e., $\partial w/\partial\theta = 0$) the equation governing the shape of the deformed interface becomes

$$d^2w/dr^2 + (1/r)dw/dr - w/\lambda^2 = 0, \qquad \text{(B4)}$$

where

$$\lambda = [T/(g\Delta\rho)]^{1/2}. \qquad \text{(B5)}$$

The solution of this equation which satisfies the boundary conditions that $w = 0$ at $r = \infty$ and $w = u$ at $r = a$ is

$$w(r) = u[K_0(r/\lambda)/K_0(a/\lambda)], \qquad \text{(B6)}$$

where K_0 is the modified Bessel function of the second kind of order zero. From Eq. B6 we can compute

$$\text{Tan}\ \phi = -dw/dr]_{r=a} = (u/\lambda)[K_1(a/\lambda)/K_0(a/\lambda)],$$

where K_1 is the modified Bessel function of the second kind of order one. Consistent with the approximations made so far we can assume $\tan\phi = \sin\phi = \phi$. Thus the force exerted by the PDMS-water interface on the edge of the poker is

$$F = 2\pi u T \Psi(a/\lambda) \qquad \text{(B7)}$$

with

$$\Psi(x) = x[K_1(x)/K_0(x)] = -1/[\gamma + \ln(x/2)] + \cdots \qquad \text{(B8)}$$

(The asymptotic approximation in Eq. B8 is valid as $x \to 0$.) The constant γ is Euler's Constant with a value of ~0.5772. From Eq. 1 of this paper we see that the long-time relaxation modulus G_∞ can be written as

$$G_\infty = F/(8au) = (\pi/4)(T/a)\Psi(a/\lambda). \qquad \text{(B9)}$$

For PDMS the surface tension is ~20 dynes/cm at 20° [2B] and ρ_g = 0.968 g/cm^3. Given that the surface tension of water at 20°C is 73 dynes/cm the interfacial tension T is <73 − 20 = 53 dynes/cm (Wu, 1982). In fact the interfacial tension of the boundary between pure PDMS and water is 42.5 dynes/cm, but impurities present in commercially available PDMS lower this value to 29–32 dynes/cm (M. Owens, Dow Chemical Corp. personal communication); we have used a value of 30 dynes/cm in the calculation below. In the experiments reported here the poker radius was 12.5 μm, so that λ = 0.978 cm, Ψ = 0.147, and

$$G_\infty = 2{,}781 \text{ dynes/cm}^2 = 278 \text{ Pa}. \qquad \text{(B10)}$$

Note that these linearized calculations give a value of G_∞ which is independent of indentation depth, and that the linearizing approximations are valid if $(u/\lambda) \ll 1$, even if $(u/a) \gg 1$. It can be easily shown that the resultant of the pressure forces acting on the flat tip of the poker is completely negligible compared with the surface tension force.

The study reported in this paper began with measurements by Bill Daily on thin layers of PDMS. His results will be reported in a subsequent publication. We are especially grateful to him for his participation in this work and his many useful contributions. We also are grateful to William B. McConnaughey whose contributions have been essential to carrying out the experiments. We also thank Drs. Anthony P. Wright and Michael Owens of Dow Chemical Corp. for information about the PDMS samples and helpful discussion.

This work was supported by National Institutes of Health grant GM38838; and by National Science Foundation grant DMB-8610636.

Received for publication 29 August 1988 and in final form 12 December 1988.

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