Mater. Res. Soc. Symp. Proc. Vol. 1049 © 2008 Materials Research Society 1049-AA06-05

Quasi-static and Oscillatory Indentation in Linear Viscoelastic Solids

Yang-Tse Cheng[1] and Che-Min Cheng[2]
[1]General Motors R&D Center, Warren, MI, 48090
[2]Institute of Mechanics, Chinese Academy of Sciences, Beijing, 100080, China, People's Republic of

ABSTRACT

Instrumented indentation is often used in the study of small-scale mechanical behavior of "soft" matters that exhibit viscoelastic behavior. A number of techniques have been used to obtain the viscoelastic properties from quasi-static or oscillatory indentations. This paper summarizes our recent findings from modeling indentation in linear viscoelastic solids. These results may help improve methods of measuring viscoelastic properties using instrumented indentation techniques.

INTRODUCTION

Instrumented indentation [1-13] can be performed in either quasi-static or oscillatory mode for measuring mechanical properties of "soft" matters, such as polymers, composites, and biomaterials, that are often viscoelastic. In the load- or displacement-controlled quasi-static mode, the load-displacement curves are recorded. One of the widely used methods, due to Oliver and Pharr [2], is to obtain the elastic modulus from the initial unloading slope, $S = (dF/dh)_m$, at the maximum indenter displacement, h_m,

$$S = \frac{dF}{dh}\Big|_{h=h_m} = \frac{4G}{1-\nu} a = \frac{2E}{\sqrt{\pi}\left(1-\nu^2\right)}\sqrt{A}\,, \tag{1}$$

where G is the shear modulus, ν is Poisson's ratio, $E = 2G(1+\nu)$ is Young's modulus, a is the contact radius, and $A = \pi a^2$ is the contact area. The contact radius, a, can be obtained from the contact depth, h_c, and indenter geometry. Oliver and Pharr [2] proposed an equation for h_c:

$$h_c = h_m - \xi \frac{F_m}{(dF/dh)_m}\,, \tag{2}$$

where F_m is the load at h_m. The numerical value of ξ is $(2/\pi)(\pi-2) = 0.727$ and $3/4$ for a conical and paraboloid of revolution, respectively. Although Eqs. (1) and (2) were derived from solutions to elastic contact problems, they have been used for indentation in elastic-plastic solids and viscoelastic solids. One of our motivations was to evaluate whether Eqs. (1) and (2) could be used for indentation in linear viscoelastic solids and another was to improve the existing methods [14-18].

In the oscillatory mode, a sinusoidal force is typically superimposed on a quasi-static load on the indenter [1,3,4,6,9,10,13]. The indentation displacement response and the out-of phase angle between the applied harmonic force and the assumed harmonic displacement may be recorded at a given excitation frequency or multiple frequencies. Several authors [1,3,4,6,9,10,13] have proposed analysis procedures for determining the complex Young's modulus, $E^*(\omega) = E'(\omega) + iE''(\omega)$, where $E'(\omega)$ is the storage modulus and $E''(\omega)$ is the loss modulus, from oscillatory indentations using the following equations:

$$\frac{E'}{1-\nu^2} = \frac{\sqrt{\pi}}{2}\frac{S}{\sqrt{A}} \quad \text{and} \quad \frac{E''}{1-\nu^2} = \frac{\sqrt{\pi}}{2}\frac{C\omega}{\sqrt{A}} \ , \tag{3}$$

where ν is Poisson's ratio, S is contact stiffness, C is damping coefficient, and A is contact area between the indenter and the sample. For an ideal indenter with infinite system stiffness and zero mass, the contact stiffness and damping coefficient are given by $S = |\Delta F / \Delta h| \cos\phi$ and $C\omega = |\Delta F / \Delta h| \sin\phi$, where ΔF is the amplitude of sinusoidal force with angular frequency ω, Δh is the amplitude of oscillatory displacement, and ϕ is the phase angle of the displacement response. Thus, by measuring displacement amplitude and phase angle under harmonic oscillation, the reduced storage and loss modulus, $E'/(1-\nu^2)$ and $E''/(1-\nu^2)$, can be obtained from

$$\frac{E'}{1-\nu^2} = \frac{\sqrt{\pi}}{2\sqrt{A}}\left|\frac{\Delta F}{\Delta h}\right|\cos\phi \quad \text{and} \quad \frac{E''}{1-\nu^2} = \frac{\sqrt{\pi}}{2\sqrt{A}}\left|\frac{\Delta F}{\Delta h}\right|\sin\phi \ . \tag{4}$$

Recently, we showed that Eq. (4) is the result of linear approximation of oscillatory indentation. By performing a nonlinear analysis, we derived the corresponding set of equations without evoking the small amplitude oscillation assumption [19].

Contact mechanics of linear viscoelastic bodies became an active area of research since the mid 1950s by the work of Lee [20], Radok [21], Lee and Radok [22], Hunter [23], Gramham [24,25], and Ting [26,27]. They have derived general equations for various contact conditions. For example, they have shown, for conical indentation in a linear viscoelastic solid with a constant Poisson's ratio, the force, $F(t)$, is given by:

$$F(t) = \frac{8\tan\theta}{\pi(1-\nu)}\int_0^t G(t-\tau)h(\tau)\frac{dh(\tau)}{d\tau}d\tau \, , \tag{5}$$

where $G(t)$ is the relaxation modulus which is related to the time-dependent Young's modulus by $E(t) = 2G(t)(1+\nu)$ and θ the half included angle of the indenter. When force is the independent variable, the displacement, $h(t)$ is given by:

$$h^2(t) = \frac{\pi(1-\nu)}{4\tan\theta}\int_0^t J_s(t-\tau)\frac{dF(\tau)}{d\tau}d\tau \, , \tag{6}$$

where $J_s(t)$ is the shear compliance. Eqs. (5) and (6) were derived based on the assumption of that the contact area is a monotonically increasing function of time. Under the same assumption, Ting and Gramham showed that the ratio of contact depth to indenter displacement is the same as that in the purely elastic case [24, 26],

$$\frac{h_c(t)}{h(t)} = \frac{2}{\pi} \, . \tag{7}$$

QUASI-STATIC INDENTATION

Conical indentation in linear viscoelastic solids: initial unloading slopes without a holding-period

We have recently shown [15] that Eqs. (5)-(7) could be used to analyze initial unloading after a loading period with a non-decreasing function $h(t)$ or $F(t)$. Specifically, the initial unloading slope is given by, using Eqs. (5)-(7),

$$\frac{dF}{dh}\bigg|_{h=h_m} = \frac{4h_c \tan\theta}{1-\nu} \frac{1}{J(0) - \dfrac{1}{v_F}\displaystyle\int_0^{t_m} \frac{dJ_s(\eta)}{d\eta}\bigg|_{\eta=t_m-\tau} \frac{dF(\tau)}{d\tau} d\tau} \ , \tag{8}$$

for load-controlled indentation with an initial unloading rate $v_F = |dF/dt|$, and

$$\frac{dF}{dh}\bigg|_{h=h_m} = \frac{4\tan\theta}{1-\nu}[G(0)h_c(t_m^+) - \frac{2}{\pi v_h}\int_0^{t_m} \frac{dG}{d\eta}\bigg|_{\eta=t_m-\tau} h(\tau)\frac{dh(\tau)}{d\tau} d\tau], \tag{9}$$

for displacement-controlled indentation with initial unloading displacement rate $v_h = |dh/dt|$. Eqs. (7-9) have been validated using finite element calculations for fast unloading after loading with a monotonically increasing function $h(t)$ or $F(t)$ [14,15]. Under fast unloading the second terms in Eqs. (8) and (9) are negligible, these equations become the same as Eq. (1) with $G(0) = 1/J_s(0)$ in place of G. Thus, the "instantaneous" properties, $G(0)/(1-\nu)$ or $E(0)/(1-\nu^2)$, can be obtained from either displacement- or load-controlled indentation measurements using Eqs. (7)-(9), provided that the unloading rate, v_h or v_F, is sufficiently fast. When unloading rates are sufficiently fast, the unloading slope is no longer a function of the unloading rate. Our finite element calculations suggested that "sufficiently fast" unloading could be achieved when the time duration of linear unloading was about 0.1 to 0.01 times the relaxation time of linear viscoelastic materials [15]. In practice, several indentation experiments with different unloading rates spanning several orders of magnitudes may be necessary to access whether unloading rates are fast enough. It is possible that required fast unloading is unachievable in practice. It is therefore convenient to develop techniques where an arbitrary unloading rate is sufficient to allow the determination of the instantaneous modulus. Methods of "load-hold-unload" discussed in the next section make such a measurement possible.

Conical indentation in linear viscoelastic solids: initial unloading slopes with a holding-period

We consider a load profile consisting of a loading period where the force is given by a monotonically increasing function, a "hold-at-the-peak load" period with a constant force, and an unloading period with an initial unloading rate, $v_F = |dF/dt|$. We have shown [18], using Eq. (8), that

$$\frac{(1-\nu)}{4a} J_s(0) = \frac{1}{dF/dh} + \frac{dh/dt\big|_{t=t_m^-}}{v_F}. \tag{10}$$

Thus, the instantaneous properties, $G(0)/(1-\nu)=1/[J_s(0)(1-\nu)]$, can be obtained from the measurement of initial unloading slope, dF/dh, the velocity of the indenter immediately before unloading, $(dh/dt)_{t=t_m^-}$, the rate of unloading, v_F, and the contact radius a. Eq. (10) was first suggested by Ngan and co-workers [12]. Eq. (10) shows that, under load-control, the "hold-at-the-peak-load" method provides a convenient means to determine the instantaneous modulus. In particular, when the holding period is sufficiently long, the ratio of $(dh/dt)_{t=t_m^-}$ over v_F becomes negligibly small as a result of creep. The instantaneous modulus can then be obtained directly from the unloading slope dF/dh and the contact radius or depth.

We have also proposed a "hold-at-the-maximum-displacement" method for indentation measurements when displacement is the independent variable [18]. We considered a displacement profile where the displacement is given by a monotonically increasing function, a hold period at a constant displacement, and an unloading period with an initial unloading rate, $v_h=|dh/dt|$. Using Eq. (9), we have shown [18] that

$$\frac{4G(0)}{1-\nu}a=\frac{dF}{dh}\bigg|_{h=h_m}+\frac{dF/dt\big|_{t_m^-}}{v_h}. \tag{11}$$

Eq. (11) shows that $G(0)/(1-\nu)$ can be obtained by measuring the initial unloading slope, dF/dh, the rate of force relaxation immediately before unloading, $(dF/dt)_{t=t_m^-}$, the rate of unloading, v_h, and the contact radius a. When the holding period is sufficiently long, the ratio of $(dF/dt)_{t=t_m^-}$ over v_h becomes negligibly small as a result of relaxation. The instantaneous modulus can be obtained directly from the unloading slope dF/dh and the contact radius or depth.

We found, using finite element calculations [15, 16,18], that the Oliver-Pharr equation for contact depth (Eq. (2)) often produces significant errors, whereas Eq. (8) is indeed a good approximation for the contact depth up to initial unloading for conical indentation. The reason that Eq. (2) is not applicable to indentation in viscoelastic solid becomes clear if we examine the two relationships used in deriving Eq. (2): $h_c/h=2/\pi$ and $h=2F/(dF/dh)$ for conical indentations. The first equation is identical to Eq. (7). However, the second equation, which comes from $F=Ch^2$ where C is a time-independent parameter, is, in general, not true for conical indentation in linear viscoelastic solids as can be seen from either Eq. (5) or (6) for the displacement- and load-controlled indentation, respectively. Thus, $E(0)/(1-\nu^2)=2G(0)/(1-\nu)$ can be obtained using Eqs. (10) or (11) together with Eq. (8) in "load-hold-unload" measurements.

OSCILLATORY INDENTATION

In oscillatory indentation measurements, a harmonic force is superimposed on a quasi-static force, i.e.,

$$F(t) = F_m f(t) + \Delta F \sin(\omega t), \tag{12}$$

where $f(t)$ is a monotonically non-decreasing function of time $|f(t)| \le 1$. Inserting Eq. (12) into (6) and using the definition of the storage and loss shear compliances, $J^{'}(\omega) = \omega \int_0^\infty J_s(s)\sin(\omega s)ds$ and $J^{''}(\omega) = -\omega \int_0^\infty J_s(s)\cos(\omega \cdot s)ds$, we obtain [19]:

$$h^2(t) = \frac{\pi(1-\nu)}{4\tan\theta}\left\{F_m \int_{-\infty}^{t} J_s(t-\tau)\frac{df(\tau)}{d\tau}d\tau + J^{'}(\omega)\Delta F \sin(\omega \cdot t) - J^{''}(\omega)\Delta F \cos(\omega \cdot t)\right\}. \tag{13}$$

Eq. (13) shows that $h^2(t)$ can be expressed as,

$$h^2(t) = B(t) + \Delta_2 h \sin(\omega t - \phi), \tag{14}$$

where $\Delta_2 h$ is the "square" amplitude of harmonic displacement and ϕ is the phase shift. Eq. (14) shows that the square of displacement, $h^2(t)$, is a sinusoidal function of time. This is different from the usual assumption that the displacement, $h(t)$, is a sinusoidal function of time based on which Eqs. (3-4) were derived. Comparing Eq. (14) with Eq. (13) and using the relationship between the complex modulus, E^*, and the complex shear compliance, J^*, i.e., $E^{'} = 2(1+\nu)\frac{J^{'}}{J^{'2} + J^{''2}}$ and $E^{''} = 2(1+\nu)\frac{J^{''}}{J^{'2} + J^{''2}}$, we obtained [19],

$$B(t) = \frac{\pi(1-\nu)}{4\tan\theta}F_m \int_{-\infty}^{t} J_s(t-\tau)\frac{df(\tau)}{d\tau}d\tau, \tag{15}$$

$$E^{'} = \frac{\pi(1-\nu^2)}{2\tan\theta}\frac{\Delta F}{\Delta_2 h}\cos\phi \quad \text{and} \quad E^{''} = \frac{\pi(1-\nu^2)}{2\tan\theta}\frac{\Delta F}{\Delta_2 h}\sin\phi. \tag{16}$$

Eq. (16) shows that the storage and loss modulus can be obtained by measuring the square amplitude of displacement $\Delta_2 h$ and phase shift ϕ. The measurement of the contact area or the absolute position of the indenter is unnecessary, thus removing the difficulties associated with contact area measurement and thermal drift for both large and small amplitude oscillatory indentations.

CONCLUSIONS

We have provided an overview of our recent studies of indentation in linear viscoelastic solids. These studies established basic equations for several methods of obtaining viscoelastic properties using quasi-static and oscillatory indentations. Specifically, we showed that the instantaneous modulus, $E(0)/(1-\nu^2) = 2G(0)/(1-\nu)$, can be obtained using either the method of fast unloading or the method of load-hold-unload for both load- and displacement-controlled quasi-static measurements. We also derived equations for obtaining the storage and loss modulus from oscillatory indentation measurements without using the usual assumption of small amplitude oscillations. Although we focused our discussions on conical indentations in this overview, corresponding equations have been derived for quasi-static and oscillatory indentations in linear viscoelastic solids using spherical indenters [16,19]. We hope these results will help improve the current practices of using indentation to determine viscoelastic properties of "soft" materials, including polymers, composites, and biomaterials.

ACKNOWLEDGMENTS

We would like to thank Dr. Wangyang Ni for his contributions to the work summarized in this paper. We would also like to thank Mike Lukitsch, Yue Qi, Tom Perry, and Mark W. Verbrugge for valuable discussions. C.-M. Cheng would like to acknowledge partial support from NSF of China, Project No.10372101.

REFERENCES

1. J. B. Pethica and W. C. Oliver, Phys. Scr. T19, 61 (1987).
2. W. C. Oliver and G. M. Pharr, J. Mat. Res. 7, 1564 (1992).
3. J. L. Loubet, B. N. Lucas, and W. C. Oliver, in International Workshop on Instrumental Indentation, San Diego, CA, April 1995, D.T. Smith (ed.), 31 (1995).
4. S. A. Syed, K. J. Wahl, and R. J. Colton, Rev. Sci. Instrum. 70, 2408 (1999).
5. S. Shimizu, T. Yanagimoto, M. Sakai, J. Mat. Res. 14, 4075 (1999).
6. N. A. Burnham, S. P. Baker, and H. M. Pollock, J. Mat. Res. 15, 2006 (2000).
7. L. Cheng *et al.*, J. Poly. Sci.: Part B: Poly. Phys. 38, 10 (2001).
8. M. L. Oyen and R. F. Cook, J. Mat. Res. 18, 139 (2003).
9. M. R. VanLandingham, J. Res. Nat. Inst. Stand. Tech. 108, 249 (2003).
10. A.C. Fischer-Cripps, Nanoindentation, 2^{nd} edition (Springer-Verlag, New York, 2004).
11. G. Huang, B. Wang and H. Lu, Mech. Time-Dependent Mater. 8, 345 (2004).
12. A.H.W. Ngan, H.T. Wang, B. Tang, K.Y. Sze, Int. J. Solids and Struct. 42, 1831 (2005).
13. G. M. Odegard, T. S. Gates and H. M. Herring, Exp. Mech. 45, 130 (2005).
14. Y.-T. Cheng and C.-M. Cheng, Mat. Sci. Eng. R44, 91 (2004).
15. Y.-T. Cheng and C.-M. Cheng, J. Mat. Res. 20, 1046 (2005).
16. Y.-T. Cheng and C.-M. Cheng, Materials Science and Engineering A 409, 93 (2005).
17. Y.-T. Cheng and C.-M. Cheng, Appl. Phys. Lett. 87, 111914 (2005).
18. Y.-T. Cheng, W. Ni, and C.-M. Cheng, J. Mat. Res. 20, 3061 (2005).
19. Y.-T. Cheng, W. Ni, and C.-M. Cheng, Phys. Rev. Lett. 97, 075506 (2006).
20. E. H. Lee, Quarterly Appl. Math. 13, 183 (1955).
21. J. R. M. Radok, Quarterly Appl. Math. 15, 198 (1957).
22. E. H. Lee, J. R. M. Radok, J. Appl. Mech. 27, 438 (1960).
23. S. C. Hunter, J. Mech. Phys. Solids 8, 219 (1960).
24. G. A. C. Graham, Int. J. Eng. Sci. 3, 27 (1965).
25. G. A. C. Graham, Int. J. Eng. Sci. 5, 495 (1967).
26. T. C. T. Ting, J. Appl. Mech. 33, 845 (1966).
27. T. C. T. Ting, J. Appl. Mech. 35, 248 (1968).