Damping Properties of Lead Metaniobate

Taeyong Lee and Roderic S. Lakes

***Abstract*—Mechanical damping, tan δ, of lead metaniobate was determined experimentally over a wide range of frequency. Damping at audio and sub-audio frequency was lower than at ultrasonic frequency. The experiments were conducted in torsion and bending using an instrument capable of determining viscoelastic properties over more than 10 decades of time and frequency. Mechanical damping was higher in bending than torsion at all frequencies. Damping observed in this study at the highest frequencies approach the high value 0.09 previously observed at ultrasonic frequency.**

I. Introduction

Viscoelasticity in transient experiments is manifested as creep (increase of strain under constant stress) or relaxation (decrease of stress under constant strain). Viscoelasticity in dynamic tests with sinusoidal loading is manifested as a phase angle δ between stress and strain; tan δ is referred to as mechanical damping and is approximately equal to the inverse of the mechanical quality factor Q. Polymers [1] often exhibit large viscoelastic effects (the loss tangent, tan δ from 0.1 to 1 or more) at ambient temperature. In structural metals [2], [3], such as steel, brass, and aluminum, viscoelastic effects are usually small (tan δ of 10^{-3} or less); some alloys exhibit a small tan $\delta < 10^{-5}$. All materials exhibit some viscoelastic response. High mechanical damping in metals is of interest in the context of damping of ship propellers [4] or in applications such as solders, involving temperatures that are a large fraction of the melting point [5]. Piezoelectric materials are also of interest in the damping of structural vibrations. A resistive circuit element can be attached to a piezoelectric inclusion in the structure or material to achieve substantial damping [6], [7], [8]. Lumped circuit elements are cumbersome; therefore, a distributed damping system would be of practical value. Damping of lead metaniobate is of interest because it does not depend on the presence of an external circuit; it is intrinsic to the material.

Lead metaniobate is a piezoelectric and ferroelectric material originally notable [9], [10] for its high Curie temperature. In ceramic form, it exhibits a Young's modulus E = 46 GPa and viscoelastic damping tan $\delta = 0.09$ in the longitudinal direction [11]. The damping is large for a ceramic and is large for a material of that stiffness. By contrast, shear moduli in other piezoelectric ceramics range from 21 to 45 GPa and tan δ from 3×10^{-3} to 0.02 for lead titanate zirconates. The high damping of lead metaniobate is useful in generating and receiving pulsed waveforms via ultrasonic transducers.

Damping of lead metaniobate has been reported only at a resonant ultrasonic frequency. The present study was conducted to explore its damping over a range of frequency.

II. Materials and Methods

Specimens of lead metaniobate were prepared from commercially available transducer disks (Valpey-Fisher Corporation, Hopkinton, MA). These were poled by the manufacturer for use in transducers. A low speed diamond wafering saw was used to cut prismatic specimens 1 to 2.4 mm across and 10 to 22 mm long. Two samples were prepared from a 2.4-cm (1-in) diameter disk in the x-direction (transverse to the disk normal) with dimensions 1.5 × 1.5 × 10.5 mm and in the z-direction (longitudinal, parallel to the disk normal) with dimensions 2.4 × 2.2 × 21.3 mm. Only a transverse sample could be obtained from the 1.2-cm (0.5-in) diameter disk because of its small thickness of 3.4 mm. Careful handling of the sample was required for test preparation because the ceramic is brittle.

Viscoelastic measurements were performed in torsion and bending at 23 ± 1°C using apparatus of Chen and Lakes [12] as modified by subsequent workers [13], [14], [15]. This device (Fig. 1) permits measurements over as much as 11 decades of time and frequency under isothermal conditions [16]. Such capability is particularly useful in the study of materials that are not thermorheologically simple. The wide frequency range is obtained by eliminating resonances from the devices used for loading and for displacement measurement, by minimizing the inertia attached to the specimen, and by use of a geometry, giving rise to a simple specimen resonance structure amenable to simple analysis. Torque was produced by the electromagnetic action of a Helmholtz coil upon a high intensity neodymium iron boron magnet at the specimen-free end. The electrical input was sinusoidal for dynamic studies and step function for creep studies. Angular displacement was measured via a split-diode light detector that measured the motion of laser light reflected from a small mirror upon the magnet. The detector signal was amplified

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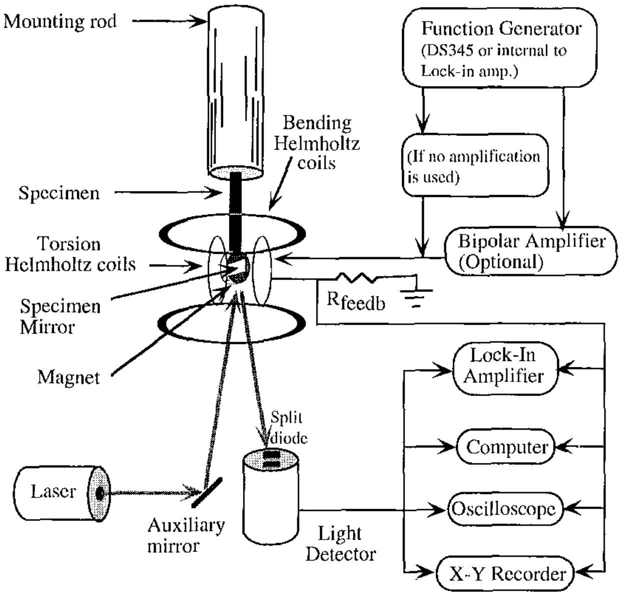


Fig. 1. Schematic diagram of the broadband viscoelastic spectroscopy apparatus.

with a wideband differential amplifier. Tan δ at resonant frequencies was inferred from the width of the dynamic compliance curve or from free decay of vibration. In the subresonant domain, $\tan\delta$ was inferred from the phase angle between torque and angle. Calibrations were performed using the well-characterized 6061-T6 aluminum alloy (G = 25.9 GPa; $\tan\delta \approx 3.6 \times 10^{-6}$) [17].

Quasistatic (creep) experiments were conducted by applying a step function current and monitoring both the current and the angular displacement signal as a function of time. Near resonances, signals were measured using a digitizing oscilloscope. At low frequency, the phase angle between torque and angular displacement was determined from the width of elliptic Lissajous figures. Creep and low frequency data were acquired via a digital data acquisition system containing a Macintosh IIci computer and LabVIEW® (Apple Computer, Cupertino, CA) interface hardware and software. At frequencies above 0.01 Hz, phase angle was determined using a lock-in amplifier (SRS 850, Stanford Research, Sunnyvale, CA) with claimed phase resolution 0.001 deg. The frequency range was segmented into regions less than 1 Hz and greater than 1 Hz because the wide frequency range necessitated different time constant settings. Load level was intentionally varied in tests of linearity at the higher frequencies. The instrument was isolated from vibration via a system of springs and viscoelastic elastomer. Low frequency noise caused by air currents was eliminated by a Plexiglas® cover.

Data reduction was conducted as follows. For frequencies sufficiently below the first natural frequency, torsional stiffness is given by $|G^*| \approx K\frac{|M^*|L}{\theta}$, and loss is given by $\tan\delta \approx \tan\phi$ in which K is a geometrical factor for torsion of a rectangular section and ϕ is the observed phase difference. For quasistatic bending, $|E^*| \approx \frac{|M^*|L}{\frac{1}{12}bh^3\theta}$, with b and h as the rectangular cross-section dimensions. In the subresonant domain, the lumped relation tan $\delta = \tan\ \phi\ (1 - (\omega/\omega_o)^2)$ was used, with ω_o as the fundamental resonance angular frequency. An exact solution is available [18] but is not needed unless $\tan\delta > 0.2$. At the resonance angular frequencies ω_o in torsion and bending, damping was calculated using the width $\Delta\omega$ at half maximum of the curve of dynamic structural compliance θ/M^* as follows:

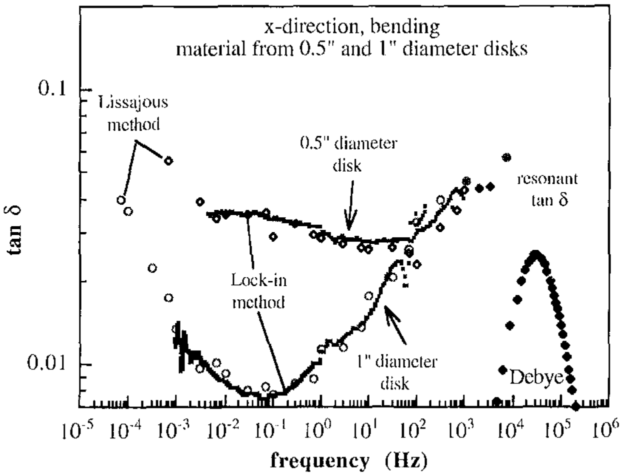


Fig. 2. $\tan\delta$ of material from 0.5- and 1-in diameter discs in x-direction, bending.

$$\tan\delta \approx \frac{1}{\sqrt{3}}\,\frac{\Delta\omega}{\omega_0}\,. \tag{1}$$

In most experiments, input voltage (and thus shear stress) was held constant. The maximum surface strain at 1 Hz was 3.18×10^{-6} for torsion and 4.61×10^{-6} for bending. To distinguish linear from nonlinear behavior, the shape of the Lissajous figures was examined, and some tests were repeated at different strain levels.

III. Results and Discussion

Results for lead metaniobate shown in Fig. 2 to 5 for both bending and torsion disclose $\tan\delta$ in the range from 0.007 to 0.1. Damping results obtained from Lissajous figures are shown as open symbols. Damping results obtained from the lock-in amplifier are shown as small solid symbols. No deviations from linearity were observed at the small strains used in this study. The instrument's capability for a wide frequency range is attained at the expense of high load capability. At large strains we would expect to observe nonlinear behavior.

Fig. 2 discloses the damping properties in bending of (transverse) x-direction specimens. Tan δ of material from 0.5- and 1-in diameter discs showed a relative minimum

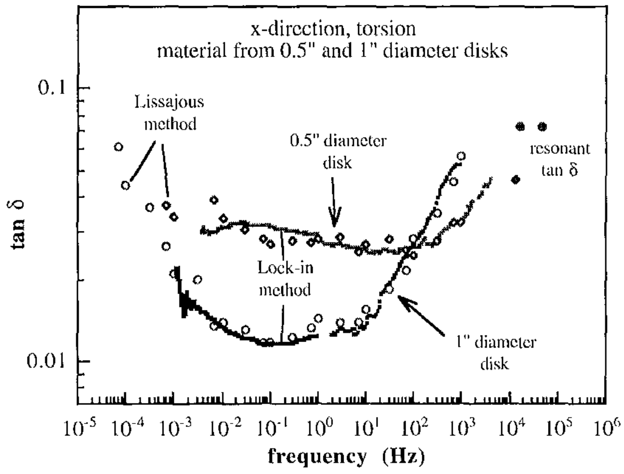


Fig. 3. tan δ of material from 0.5- and 1-in diameter discs in x-direction, torsion.

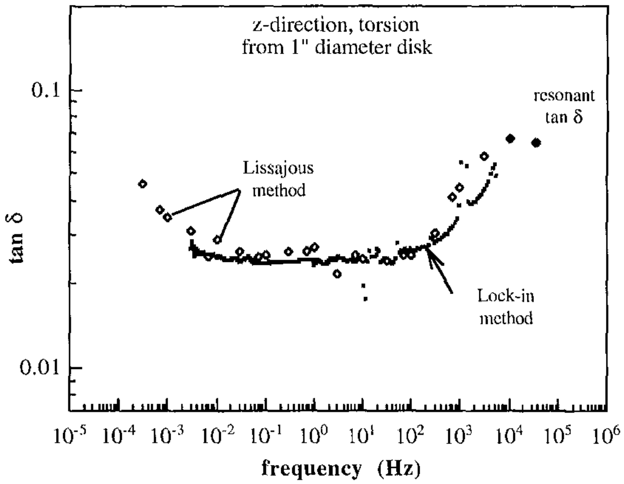


Fig. 5. tan δ of material from 1-in diameter discs in z-direction, torsion.

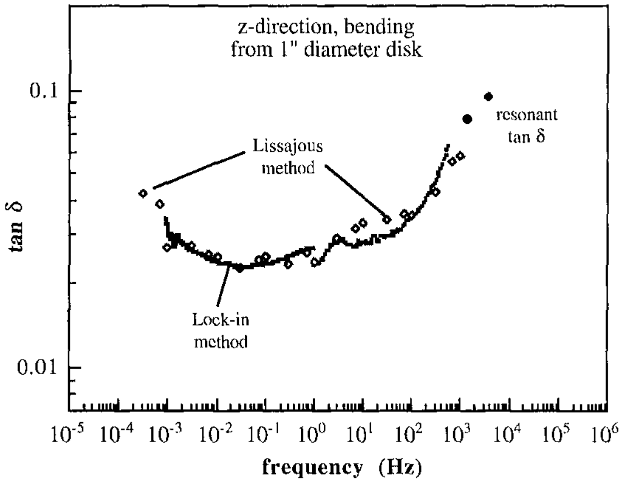


Fig. 4. tan δ of material from 1-in diameter discs in z-direction, bending.

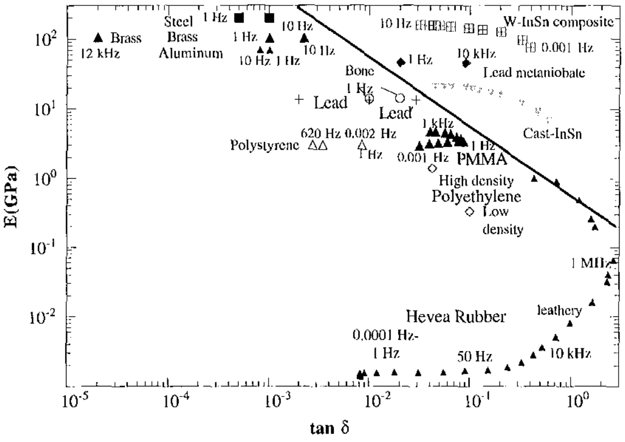


Fig. 6. Stiffness loss map showing comparison of lead metaniobate properties with those of other materials.

at sub-audio to low audio frequency. A similar minimum occurred in torsion for the x-direction as well, as seen in Fig. 3. The sample from the 0.5-in diameter disc showed similar tan δ in both bending and torsion tests, but showed higher tan δ at resonances in torsion. Overall damping in torsion was higher than in bending. Fig. 4 and 5 disclose tan δ of material from 1-in diameter lead metaniobate in the (longitudinal) z-direction. In the bending test, as seen in Fig. 4, higher tan δ at resonances was observed than in torsion. Also, mechanical damping was higher in bending than torsion at all frequencies. However, the torsion studies disclosed higher tan δ in frequencies above 100 Hz as seen in Fig. 3 and 5. Tan δ below 100 Hz was higher in the bending test. Damping values differed in the x and z directions. Anisotropy is expected in a polarized, polycrystalline sample. Moreover, samples from different size transducer disks exhibited different damping, presumably as a result of differences in the pressing and sintering process. Damping observed in this study at the highest frequencies approaches the high value 0.09 previously observed at ultrasonic frequency.

Piezoelectric materials may be of use in the damping of structural vibrations. Damping of lead metaniobate is of interest in that context because it is intrinsic to the material and because the material is relatively stiff. Properties of lead metaniobate are compared with other materials [19] in the stiffness-loss map in Fig. 6. E tan δ is a figure of merit for vibration absorption. The diagonal line in Fig. 6 presents the largest product (E tan $\delta \approx 0.6$ GPa) of stiffness E and damping found in common materials, including polymers through the glass-rubber transition and soft metals such as Pb. Stiffness is considered as the absolute value of the complex dynamic Young's modulus $|E^*|$. It is possible to achieve higher E tan δ in composites designed to achieve such a figure of merit (for vibration absorption), as done by Brodt and Lakes [20], but such a combination is unusual in materials other than designed composites. Lead metaniobate has a reasonably high product E $\tan\delta$, particularly at the higher acoustic frequencies. It could be used as a constituent in high damping composite materials.

As for transducer applications, the present results inform the designer that lead metaniobate has a lower $\tan\delta$ at acoustic frequencies, which may be encountered in bimorphs or other bender elements, than it does at ultrasonic frequencies used in determining the 'book value' of $\tan\delta$.

As for causal mechanisms, some processes such as dielectric relaxation caused by point defects in ferroelectrics give rise to a Debye form [21]. The Debye model for relaxation in the time domain is

$$E(t) = E_2 + E_1 e^{-t/\tau_r} . \tag{2}$$

The corresponding Debye peak in $\tan\delta$ in the frequency domain covers about one decade and is as follows. The frequency dependence is shown in Fig. 2 for comparison with the behavior of lead metaniobate.

$$\tan\delta(\omega) = \frac{\Delta}{\sqrt{1+\Delta}} \, \frac{\omega\tau_m}{1+\omega^2\tau_m^2} \tag{3}$$

where $\tau_m = \tau_r\sqrt{1+\Delta}$ is a time constant, $\omega = 2\pi\nu$ is angular frequency, and ν is frequency. The relaxation strength Δ is defined as the change in stiffness during relaxation divided by the stiffness at long time. Clearly, the observed damping covers a much broader range of frequency than a Debye peak; therefore, simple mechanisms that give rise to such behavior cannot play a major role. Mechanical damping in ferroelectrics and ferromagnetics can occur as a result of drag because of interaction between stress-induced domain wall motion and defects [22], including dislocations [23] and point defects [24] such as vacancies [25]. Interaction between domain walls and point defects can also give rise to dielectric loss [26] in multiple relaxation mechanisms that are operative in ferroelectrics. In temperature-dependent dielectric relaxation, low frequency dispersion is believed to originate from domain wall relaxations rather than heat diffusion [27]. The present study of dependence of $\tan\delta$ on frequency ν shows the domain wall drag cannot be simply viscous; if it were, $\tan\delta \propto \nu$. Moreover, the damping is not of the hysteresis type; if it were, $\tan\delta$ would be independent of frequency. The attainment of a complete understanding of the causal mechanisms for damping in ferroelectrics is a subject of future study.

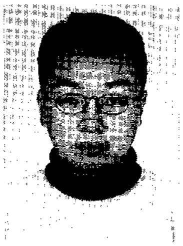
IV. CONCLUSIONS

Mechanical damping of lead metaniobate was observed to be lower in the audio frequency range than at ultrasonic frequency. The present results at the lower ultrasonic frequencies are consistent with the accepted values of damping for lead metaniobate.

REFERENCES

[1] J. D. Ferry, *Viscoelastic Properties of Polymers.* 2nd ed. New York: John Wiley, 1979.
[2] C. Zener, *Elasticity and Anelasticity of Metals.* Chicago, IL: University of Chicago Press, 1948.
[3] A. S. Nowick and B. S. Berry, *Anelastic Relaxation in Crystalline Solids.* New York: Academic Press, 1972, pp. 435–462.
[4] I. G. Ritchie, K. W. Sprungmann, and M. Sahoo, "Internal friction in Sonoston--A high damping Mn/Cu-based alloy for marine propeller applications," *J. Phys.*, vol. 46, pp. C10-409–C10-412, 1985.
[5] T. S. Kê, "Internal friction of metals at very high temperatures," *J. Appl. Phys.*, vol. 21, pp. 414–419, 1950.
[6] R. L. Forward, "Electronic damping of vibrations in optical structures," *J. Appl. Opt.*, vol. 18, pp. 690–697, 1979.
[7] R. L. Forward and C. J. Swigert, "Electronic damping of orthogonal bending modes in a cylindrical mast-Theory," *J. Spacecraft Rockets*, vol. 18, pp. 5–10, 1981.
[8] N. W. Hagood and A. von Flotow, "Damping of structural vibrations with piezoelectric materials and passive electrical networks," *J. Sound Vibration*, vol. 146, pp. 243–268, 1991.
[9] G. Goodman, "Ferroelectric properties of lead metaniobate," *J. Amer. Ceram. Soc.*, vol. 36, pp. 368–372, 1953.
[10] —– , "Ceramic composition," U.S. Patent 2805165, 1957.
[11] D. A. Berlincourt, D. R. Curran, and H. Jaffe, "Piezoelectric and piezomagnetic materials and their function in transducers," in *Physical Acoustics.* vol. 1A, E. P. Mason, Ed. 1964, pp. 169–270.
[12] C. P. Chen and R. S. Lakes, "Apparatus for determining the properties of materials over ten decades of frequency and time," *J. Rheol.*, vol. 8, pp. 1231–1249, 1989.
[13] M. Brodt, L. S. Cook, and R. S. Lakes, "Apparatus for measuring viscoelastic properties over ten decades: Refinements," *Rev. Sci. Instrum.*, vol. 66, pp. 5292–5297, 1995.
[14] J. Quackenbush, "Improvements to a method of measuring viscoelastic characteristics of solids," M.S. Thesis, University of Iowa, 1995.
[15] E. B. Garner, R. S. Lakes, T. Lee, C. Swan, and R. Brand, "Viscoelastic dissipation in compact bone: Implications for stress-induced fluid flow in bone," *J. Biomech. Eng.*, vol. 122, pp. 166–172, 2000.
[16] R. S. Lakes and J. Quackenbush, "Viscoelastic behaviour in indium tin alloys over a wide range of frequency and time," *Philosoph. Mag. Lett.*, vol. 74, pp. 227–232, 1996.
[17] W. Duffy, "Acoustic quality factor of aluminum alloys from 50 mK to 300K," *J. Appl. Phys.*, vol. 68, pp. 5601–5609, 1990.
[18] W. G. Gottenberg and R. M. Christensen, "An experiment for determination of the mechanical property in shear for a linear, isotropic viscoelastic solid," *Int. J. Eng. Sci.*, vol. 2, pp. 45–56, 1964.
[19] R. S. Lakes, *Viscoelastic Solids.* Boca Raton, FL: CRC Press, 1998.
[20] M. Brodt and R. S. Lakes, "Composite materials which exhibit high stiffness and high viscoelastic damping," *J. Composite Mater.*, vol. 29, pp. 1823–1833, 1995.
[21] M. E. Lines and A. M. Glass, *Principles and Applications of Ferroelectrics and Related Materials.* Oxford, England: Oxford University Press, 1997.
[22] D. Wang, Y. Fotinich, and G. P. Carman, "Influence of temperature on the electromechanical and fatigue behavior of piezoelectric ceramics," *J. Appl. Phys.*, vol. 83, no. 10, pp. 5342–5350, 1998.
[23] A. Wolfenden, "Internal friction in nickel near the Curie temperature," *Scripta Metall.*, vol. 12, pp. 103–109, 1978.
[24] V. S. Postnilov, V. S. Pavlov, S. A. Gridnev, and S. K. Turkov, "Interaction between 90° domain walls and point defects of the crystal lattice in ferroelectric ceramics," *Sov. Phys. Solid State*, vol. 10, pp. 1267–1270, 1968.
[25] B. L. Cheng, M. Gabbay, and G. Fantozzi, "Anelastic relaxation associated with the motion of domain walls in barium titanate ceramics," *J. Mater. Sci.*, vol. 31, pp. 4141–4147, 1996.
[26] J. O. Gentner, P. Gerthsen, N. A. Schmidt, and R. E. Send, "Dielectric losses in ferroelectric ceramics produced by domain wall motion," *J. Appl. Phys.*, vol. 49, pp. 4485–4489, 1978.
[27] B. G. Kim and J. J. Kim, "Origin of low-frequency dielectric dispersion in KH2PO4 and RbH2PO4 ferroelectric crystals," *Phys.*

Rev. B-Condensed Matter, vol. 59, no. 21, pp. 13509–13512, 1999.



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