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Kurt M. Rittenmyer and Pieter S. Dubbelday

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Underwater Sound Reference Detachment
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Orlando, FL 32856-8337

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Direct measurement of the temperature-dependent piezoelectric coefficients of composite materials by laser Doppler vibrometry

Kurt M. Rittenmyer and Pieter S. Dubbelday
Naval Research Laboratory, Underwater Sound Reference Detachment, P.O. Box 568337, Orlando, Florida 32856-8337

(Received 30 May 1991; accepted for publication 30 December 1991)

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INTRODUCTION

Measurements of piezoelectric constants can be made by a variety of techniques including the resonance method outlined in the IEEE Standard[1] which determines resonator parameters at a single frequency. For piezoelectric composite materials, which are viscoelastic, the dielectric and elastic properties vary widely over broad ranges of frequency and temperature as described in an earlier article.[2] This variation is attributed to the glass–rubber transition of the polymer material that causes softening of the elastic and dielectric stiffnesses as the temperature is increased. The influence of the glass transition on the piezoelectric coefficients of these materials is complicated and has not been fully studied. To perform measurements of the piezoelectric properties over ranges of several decades in frequency, nonresonance measurement techniques must be used. Measuring piezoelectric properties over a wide range of temperature requires instrumentation that is either designed specifically with low thermal expansion materials for this purpose, or noncontact methods, where the experimental apparatus can be physically separated from the test environment. For the latter case, optical methods of measuring acoustic displacements generated by applying an electric field (the converse piezoelectric effect) are appropriate.

Formally, the converse piezoelectric tensor d_{ijm} relates the elastic strain tensor, defined for the displacement gradient $\phi_{i,j}\,(=\partial\phi_i/\partial x_j)$ as

$$S_{ij} = \tfrac{1}{2}(\phi_{i,j} + \phi_{j,i}) \qquad (1)$$

to the electric field vector E_m in the equation

$$S_{ij} = d_{ijm}E_m, \qquad (2)$$

which defines the piezoelectric tensor d_{ijm} under boundary conditions of constant stress T_{ij} and electric field E_m. If the tensor is reduced to matrix notation by the usual convention, then for most piezoelectric ceramics and composite materials, the transposed piezoelectric matrix d_{ij} (where indices $i = 1$–3 and $j = 1$–6) has the form,

$$d_{ij} = \begin{pmatrix} 0 & 0 & d_{31} \\ 0 & 0 & d_{31} \\ 0 & 0 & d_{33} \\ 0 & d_{15} & 0 \\ d_{15} & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix} \qquad (3)$$

as only d_{33}, d_{31} ($=d_{32}$), and d_{15} ($=d_{24}$) constants are independent and nonzero. In this article, a laser Doppler velocimeter is used to measure the longitudinal (d_{33}) and lateral (d_{31}) piezoelectric coefficients from temperatures of -50 °C to 50 °C and over a frequency range from 100 Hz to 10 kHz. Since the oscillatory displacements of the surfaces of the piezoelectric samples are small, being of the order of 1–10 Å in comparison with the optical wavelength (6328 Å for a He–Ne laser), the Doppler shift is a small fraction of a wavelength. Therefore, the method is similar in principle to optical interferometry.

I. EXPERIMENTAL METHOD

The frequency of laser light from a stationary source impinging on a moving surface is shifted upon reflection according to the Doppler shift equation as shown in Fig. 1:

$$\Delta\nu = 2v\cos\theta/\lambda, \qquad (4)$$

where $\Delta\nu$ is the frequency shift of the light, v is the reflecting surface velocity amplitude, θ is the angle between the incident and reflected waves, and λ is the wavelength of the laser light. The Doppler frequency shift is proportional to the velocity of the surface. For small harmonically varying velocity, $\Delta\nu/\nu \ll 1$ and the output signal is essentially a phase-modulated signal of frequency ν. For normal incidence, the constant of proportionality in Eq. (4) depends only on the

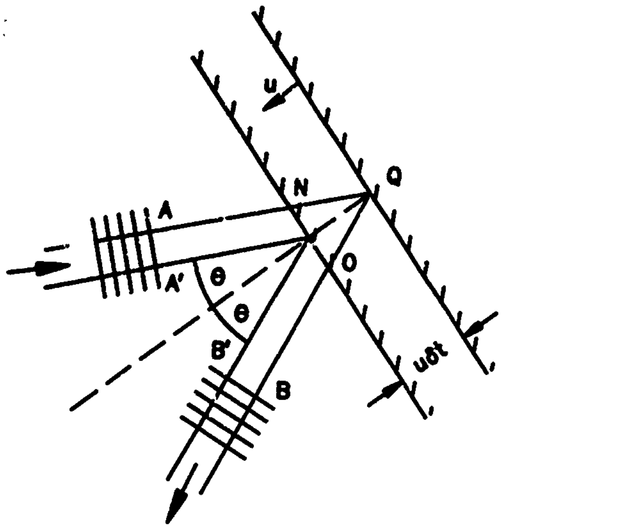


FIG. 1. Illustration of laser Doppler effect from surface (represented by hatched lines) moving with velocity v. Wavefront B–B' reflected at time t travels distance NO + NQ further than at time $t + \delta t$ creating a difference in frequency or phase between impinging wave A–A' and reflected wave B–B'.

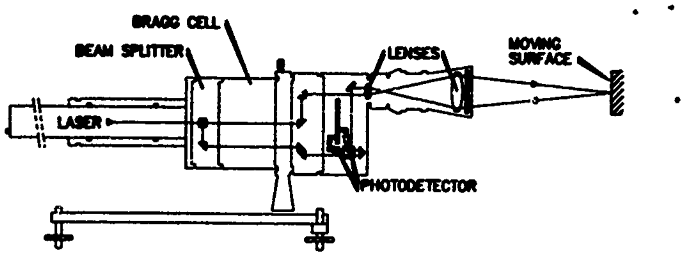


FIG. 2. Diagram of optical portion of Dantec model 55X laser Doppler vibrometer.

wavelength of the laser light.

For harmonic displacement of the form

$$\phi = \phi_0 e^{-i\omega t}, \quad (5)$$

the velocity of the surface v is described by

$$v = v_0 e^{-i\omega t} \quad (6)$$

and is related to its displacement ϕ, by

$$v = \frac{d\phi}{dt} = -i\omega\phi, \quad (7)$$

where ϕ_0 is the displacement amplitude, v_0 is the velocity amplitude, and ω is the frequency of oscillation. Using Eqs. (1) and (2), the following relationship between velocity amplitude and measured piezoelectric effect can be derived for the extensional vibrations in the thickness direction, when the thickness h is much smaller than the acoustic wavelength:

$$d_{33} = 2|v_0|/i\omega V_3, \quad (8)$$

where V_3 is the voltage applied to the sample. The d_{31} coefficient is determined by a similar relationship,

$$d_{31} = 2h\,|v_0|/i\sigma\omega V_3, \quad (9)$$

where σ is the sample diameter.

A. Principle and design of the laser Doppler velocimeter

The laser Doppler effect was first used to measure flow velocities by Yeh and Cummins.[3] The technique is reviewed in detail in the textbooks by Drain[4] and Durrani[5] and will be discussed briefly. Two laser beams intersect at the Si PIN-diode photodetectors as shown in Fig. 2. One coherent laser beam is backscattered from the vibrating surface of the sample being measured. The other beam is a reference beam which is shifted slightly in frequency from the reflected beam by using an acousto-optic Bragg cell. The reflected and reference beams are focused into a small spot at the surface of the photodiode, which produces voltage proportional to the intensity of light focused on it. The two beams produce an interference pattern at the surface of the detector. If the instrument is properly aligned, the constructive portion of the fringe covers the photodetector when the surface of the sample is at rest. The exposed area of the detector should be smaller than the fringe width so that only one-half of an interference fringe covers the detector and variations in intensity are not averaged over the surface of the detector. Motion of the surface changes the fringe spacing and therefore changes the intensity of light impinging on the photodetector. The scattering surface of the sample produces speckle. The position of the instrument is adjusted so that it is focused on a single speckle and the light that impinges on the photodiode is essentially coherent. The other signal, the reference, is shifted in frequency by 40 MHz using an acousto-optic Bragg cell and is then diffracted onto the detector by several prisms. This frequency shift causes the fringe pattern to oscillate in a direction perpendicular to the fringes at a frequency of 40 MHz. It therefore provides directional characteristics to the Doppler shift. Motion of the reflecting surface in opposition to the fringe motion increases the Doppler frequency whereas motion in the same direction decreases it. Since the photodetector is nonlinear with respect to signal amplitudes, the two laser beams will produce outputs that include the sum and difference frequencies of the two laser beams. The reflected beam is Doppler shifted according to Eq. (1). Thus the difference frequency is equal to the Doppler frequency. The instantaneous difference frequency will be equal to 40 MHz plus or minus the instantaneous Doppler frequency. The sign of the frequency shift depends on the velocity direction. The motion of the vibrating surface modulates the position, or phase of the interference fringe and therefore modulates the carrier frequency at the frequency of the vibration. A phase-locked voltage-controlled oscillator can then be used to demodulate the signal so that the signal amplitude is proportional to the Doppler velocity. Following demodulation, low-frequency signals can be accurately measured using standard phase-locking techniques.

B. Experimental arrangement

A diagram of the system used to measure piezoelectric strain is shown in Fig. 3. A variety of optical configurations for observing laser Doppler effects are possible.[4,5] For these experiments, a reference beam heterodyne LDV system (Dantec model 55X laser Doppler vibrometer) was used.

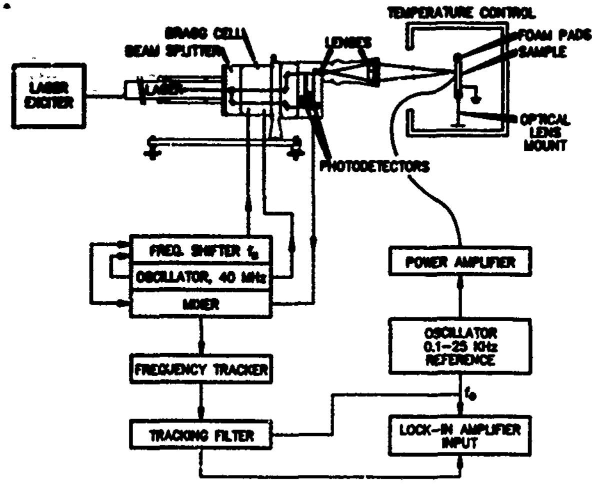


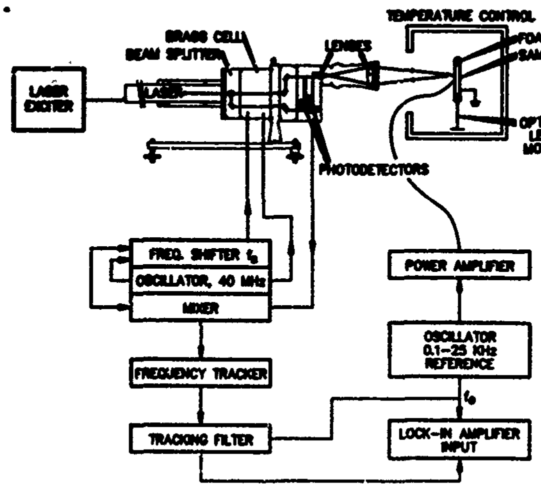
FIG. 3. Experimental apparatus for measuring temperature-dependent piezoelectric coefficients using the laser Doppler vibrometer.

This system includes the He–Ne laser source ($\lambda = 6328$ Å), beamsplitter, acousto-optic Bragg cell, Si PIN diode photodetectors, and the focusing optics. A Dantec model 55N10 frequency shifter module is used to operate the Bragg cell at 40 MHz.[6] It is capable of shifting frequencies in several ranges between 10 kHz and 9 MHz. A Dantec model 55N20 frequency tracker demodulates the signal from the photodetectors using voltage-controlled oscillators and phase-locking techniques. This signal is then passed through a tracking filter (Spectral Dynamics Corp. model SD101B dynamic analyzer) that is phase locked to the oscillator (Racal-Dana model 9085 low distortion oscillator) driving the sample through an audio power amplifier (Optimation model 12C). The output of the tracking filter is measured using a lock-in amplifier (Princeton Applied Research, Inc. model 5204). The tracking filter prevents the lock-in amplifier from overloading as the imperfectly demodulated signal still contains carrier frequency components. The electronic system as a whole is limited to measuring frequencies below 100 kHz because of the frequency response of the lock-in analyzer and the audio amplifier. This range could be extended by replacing these two components with instruments having superior frequency response. However, they are adequate for the measurements described here since the mechanical response is limited at high frequency by resonances of the sample and the support apparatus.

C. Procedure for measuring piezoelectric d_{33} and d_{31} coefficients as functions of temperature and frequency

The d_{33} and d_{31} coefficients of a piezoelectric composite material, NTK PR-306 Piezorubber, were measured using the instrumentation described. This material is a 0-3 composite with piezoelectric lead titanate particles embedded in a polychloroprene rubber matrix in roughly a 65/35 volume ratio. NTK PR-307 is a similar material, but the particles are roughly a factor of 5 larger. The piezoelectric coefficients of a Navy type I piezoelectric lead zirconate–titanate sample were also measured in order to test the effectiveness of the instrument and the sample mounting. Samples with diameters of 2–5 cm are mounted on soft, highly porous foam rubber pads in a standard 2-in.-diam optical lens mount. The mounting fixture can easily be rotated 180°. By measuring the displacement of both faces, and computing the average, flexural motions of the surfaces can be separated from pure dilatation. The foam rubber pads provide adequate isolation from vibration and allow the edges of the sample to remain essentially free of stress. Measurement of the d_{31} coefficient is made by focusing the beam on the edge of the sample disk, or alternatively, on the edge of a thin bar of the material and measuring the particle velocity as a function of voltage. Because of its high degree of porosity, the foam material maintains rather high compliance at temperatures below -30 °C. The NTK PR-306 Piezorubber disk was approximately 29 mm in diameter and 3 mm thick and was supplied with approximately 0.5 mm of carbon-filled rubber electrodes on each of the major parallel faces. The NTK PR-307 sample was 5.1 cm in diameter and was electroded with a silver paint. Very fine silver wire leads were attached to the electrodes using conductive epoxy. A small dot of reflective tape (Scotchlite) was placed at the center of the disk. The tape increases the intensity of the reflected signal, improves the signal quality, and makes alignment and focusing of the optics easier. For measuring d_{33}, the laser beam was focused on the reflective tape that was placed at the center of the face of the disk. For d_{31} measurements, the beam was focused on the side of the disk (90° incidence). Once the sample was mounted, it was placed in a liquid nitrogen-cooled environmental test chamber equipped with a transparent window. The window does not influence the measurement since the Doppler frequency is measured at the sample surface and is unaltered after it passes through the window. The sample leads were connected to the power amplifier. The laser was aligned and focused on a single speckle. This was accomplished by adjusting the instrument for the largest Doppler signal with the least noise as observed on an oscilloscope. A signal of 10 to 200 V was applied to the piezoelectric sample. The frequency of this signal was varied in the range between 100 Hz and 100 kHz. The high end of this range was altered according to the position of the lateral resonance frequency.

The sample driving-frequency was adjusted on the oscillator. The Doppler frequency range was set accordingly for the frequency shifter and the frequency tracker. The settings on these instruments trade sensitivity for frequency bandwidth. High sensitivity is required for the small piezoelectrically generated velocities encountered in these experiments. Large bandwidth is required for high-frequency measurements. For small vibration velocities, the setting of the Doppler frequency range is not critical, provided the sample vibration frequency and the Doppler frequency are not too near each other. As a general rule, the Doppler frequency generally should be at least one decade in frequency higher than the signal applied to the sample for satisfactory demodulation. For large surface velocities, care must be taken not to exceed the maximum allowable signal slew rates for a given frequency range. The amplitude of the demodulated signal is then measured using a lock-in amplifier and is directly proportional to the velocity of the sample surface.



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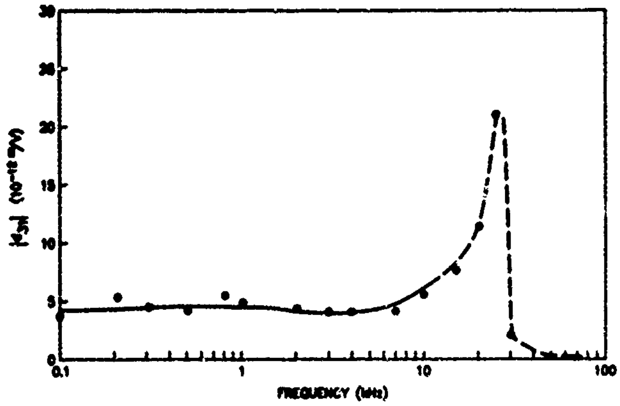

FIG. 6. Ratio of the strain amplitude in the radial direction to the applied electric field in the thickness direction as a function of driving signal frequency for a 2.5-cm disk of NTK PR-306. The solid line indicates where the influence of the resonance on the strain is negligible. The dashed line indicates where the electromechanical resonance contributes substantially to the measured value. (Lines through the data are guides to the eye.)

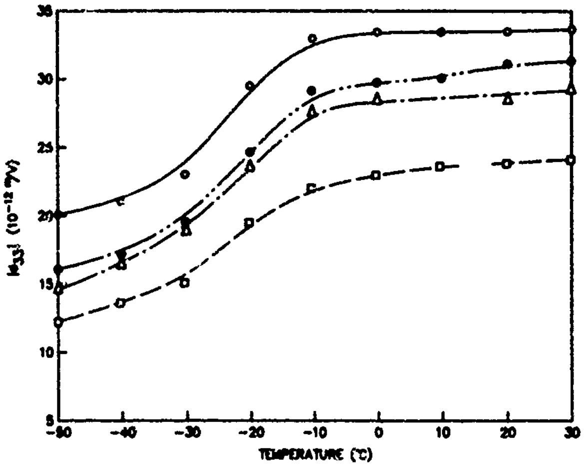

FIG. 8. Piezoelectric d_{33} coefficient as a function of temperature for NTK PR-306 Piezorubber. The following symbols denote the frequency at which the measurement was performed: O—0.4 kHz, Δ—1 kHz, □—2 kHz, ●—4 kHz

shown in Fig. 7. It is independent of frequency up to almost 10 kHz with a value of 52×10^{-12} m/V.

The d_{33} coefficient of NTK PR-306 is shown as a function of temperature in Fig. 8 for several frequencies. The influence of the glass transition below -10 °C is apparent. The value at 4 kHz increases about 40% as the temperature rises from -50 °C to 0 °C. The results of the measurements for d_{31} are plotted in Fig. 9. The values for all frequencies show a steady increase of $|d_{31}|$ with temperature starting at 1×10^{-12} m/V at -50 °C and increasing to 6×10^{-12} m/V at 40 °C. The curve is similar in form to the d_{33} measurement between -50 °C and 10 °C. Above 10 °C, the magnitude of d_{31} continues to increase whereas d_{33} approaches a constant value. Furthermore, d_{31} depends only slightly on frequency. The radial mode of the disk influences the value of the measured d_{33} coefficient. Because of the large damping for this mode, its influence extends over broad ranges of temperature and frequency and causes partial clamping of the strain in the thickness direction. This reduces the measured d_{33} value at 1 and 2 kHz. The lowest frequency measurement (0.4 Hz) is likely to be the closest to the true material constant. The higher frequency measurement (4 kHz) is slightly lower due the clamping of the material in the lateral direction. However, the effect of clamping on the radial resonance for high-frequency measurements is small because the lateral coupling is only on the order of 1%. The measured d_{33} coefficient of NTK PR-307 was quite independent of frequency below resonance. Apparently, the low lateral coupling of the material combined with the larger diameter of the sample eliminated the influence of the lateral 3-1 mode on the measurement of d_{33}.

The temperature dependence of d_{31} could be related to the presence of low-frequency flexural modes that are excit-

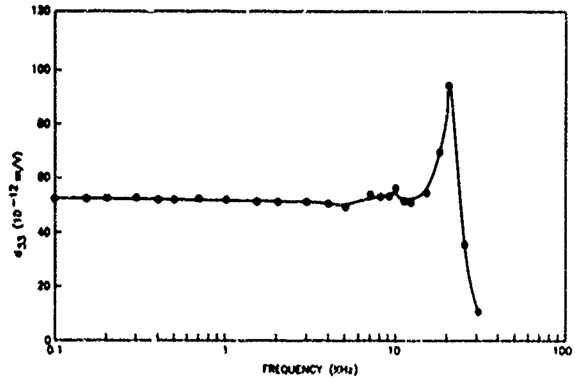

FIG. 7 Ratio of strain amplitude in thickness direction to applied electric field as a function of frequency for a 5-cm disk of NTK PR-307 The solid line indicates where the influence of the resonance on the strain is negligible The dashed line indicates where the electromechanical resonance contributes substantially to the measured value (Lines through the data are guides to the eye)

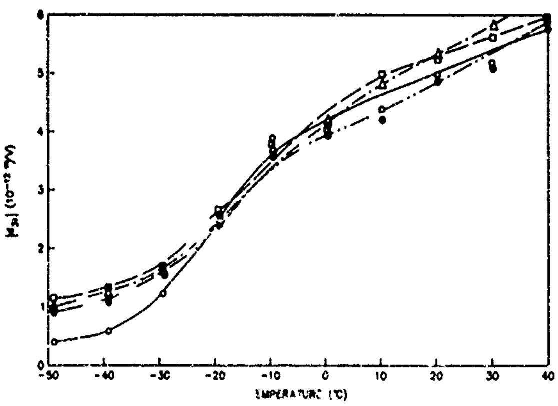

FIG 9 Piezoelectric d_{31} coefficient as a function of temperature for NTK PR 306 Piezorubber The following symbols denote the frequency at which the measurement was performed. O —0.4 kHz, Δ—1 kHz, □—2 kHz, ●—4 kHz

ed because the edges of the sample are not perfectly free. The low shear modulus of the composite material accentuates flexural motion that affects the piezoelectric response primarily through the d_{31} coefficient. The variation with frequency may possibly be explained by the presence of Lamb waves in the sample disk. The disk was as small as is reasonable for proper alignment, but may still be large enough to support waves of this type. Although care was taken to minimize flexural waves by keeping the edges of the sample as free as possible, it is not possible to eliminate them entirely. To test for their presence, measurements were made from each side of the sample at room temperature. The d_{33} coefficient was then computed from the average of the measurements thereby separating the flexural contribution from the longitudinal strain in the sample. No significant difference in d_{33} value from the first measurement was observed. Unfortunately, making the differential measurements at all temperatures is difficult without a dual beam apparatus. Therefore, the possibility of flexural contributions to the motion cannot be ruled out at all temperatures and frequencies that were measured.

The dielectric permittivity was measured at the same frequencies and temperatures as the piezoelectric constants. The dielectric constant, $(\epsilon_{33}^T/\epsilon_0)$, is plotted in Fig. 10 for frequencies of 0.4, 1, and 4 kHz. It increases roughly 40% as the temperature is increased from $-50\,^\circ$C to $0\,^\circ$C. The dielectric loss is plotted in Fig. 11. The dielectric constant was used to calculate the g_{33} value which is plotted as a function of temperature at several frequencies in Fig. 12.

The hydrostatic coefficient d_h, is shown as a function of temperature in Fig. 13. There are two sets of results. One set is measured directly using the reciprocity coupler technique. The other set is calculated using Eq. (8) and the measured values of d_{33} and d_{31}. Similar results are shown for the g_h coefficient.

III. DISCUSSION

The laser Doppler technique was applied to the measurements of piezoelectric coefficients of ceramic and composite materials as demonstrated by the measurements of

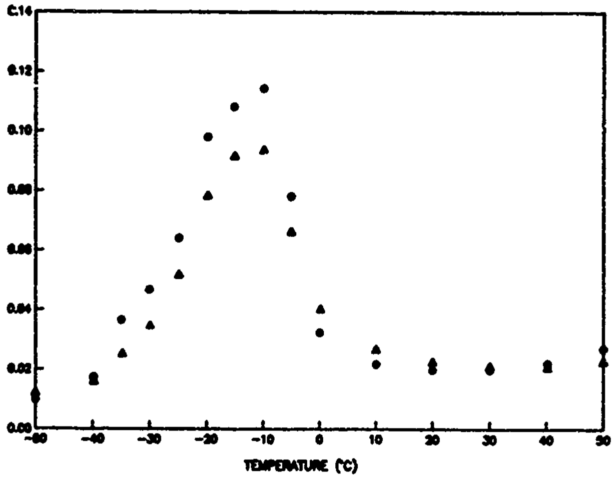

FIG. 11. Dielectric loss tangent as a function of temperature. (Δ—1 kHz, ●—4 kHz)

both d_{33} and d_{31} of lead zirconate–titanate and piezoelectric composite materials at frequencies up to 60 kHz. For the composite material, the measurements show that the magnitude of the piezoelectric coefficient is readily determined. Measurement of the complex part is difficult and was not attempted.

The results of measurements on the composite material indicate that the glass–rubber transition largely influences the piezoelectric d_{33} coefficient through the dielectric permittivity. If this were absolutely true, the percent change in d_{33} would be equal to the percent change in ϵ_{33}^T. The g_{33} coefficient, which is defined as

$$g_{33} = d_{33}/\epsilon_{33}^T, \tag{11}$$

would then be nearly independent of temperature. Figure 12 shows that g_{33} increases roughly 15% between $-50\,^\circ$C and $+30\,^\circ$C. The d_{33} coefficient increases approximately 45% over the same range. Therefore, approximately two-thirds of the temperature dependence of the d_{33} coefficient may be attributed to the dielectric constant. The remainder is relat-

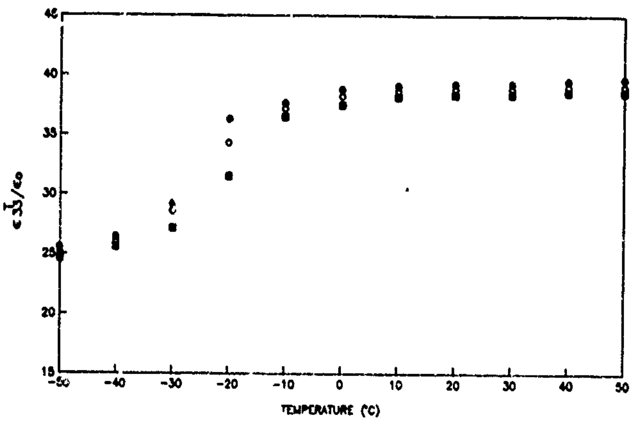

FIG 10. Relative-dielectric permittivity of NTK PR-306 as a function of temperature. (Frequency of measurement □—0.4 kHz, ○—1 kHz, ●—4 kHz)

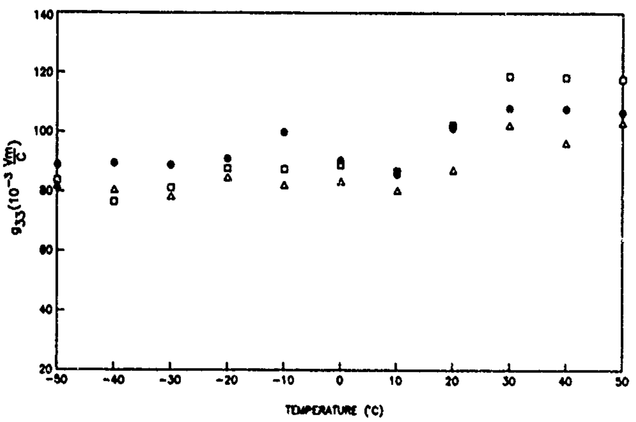

FIG. 12 Piezoelectric g_{33} coefficient of NTK PR-306 Piezorubber as a function of temperature The following symbols denote the frequency at which the measurement was performed. ○—0.4 kHz, Δ—1.0 kHz, □—2 kHz, ●—4 kHz

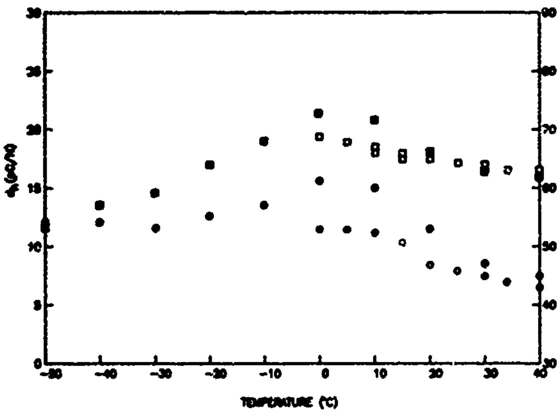

FIG. 13. Hydrostatic piezoelectric coefficients d_h (□) and g_h (○) of NTK PR-306 Piezorubber as a function of temperature. Filled symbols represent values calculated from direct laser Doppler measurements using Eqs. (10) and (11). Open symbols represent direct measurements of g_h and d_h. All measurements are at 1 kHz.

ed to the elastic compliance of the polymer which changes by more than an order of magnitude in the glass transition region.[2] Some motion of the sample in the holder is unavoidable, and despite the precautions mentioned earlier, may change with frequency thus causing the measured d_{33} values to vary with frequency. However, since the results were quite repeatable, it is doubtful that the exact alignment of the sample in the holder affects the measurement to such an extent. The magnitude of d_{33} and real part of ϵ_{33}^T were used in calculating g_{33}. The dielectric loss only becomes significant below 10 °C and is always less than 0.2. Therefore, neglecting the dielectric loss results in less than a 4% error in the calculation of g_{33}. The imaginary part of d_{33} was not determined.

The measured values of d_h are reasonably consistent with the values calculated from the d_{33} and d_{31} measurements for temperatures above 0 °C. The frequency of the measurements is 1 kHz. The apparatus used for the d_h measurement is limited to temperatures above − 10 °C and cannot operate in the glass transition region. However, Fig. 12 shows that the values match each other fairly well over the temperatures where the direct measurements and the calculated values overlap. Moisture absorption in the sample affects the dielectric properties for temperatures near 0 °C. This problem is unavoidable in the laser measurements. The reciprocity coupler measurements, however, are performed in degassed castor oil and are free of moisture contamination. This may account for the rather small differences between the optical measurements and the acoustic measurements at temperatures near 0 °C.

One would think that the softer polymer with low shear stiffness at high temperature would provide better isolation between the longitudinal and transverse motion resulting in lower values of d_{31}. The opposite behavior is observed. This is due, in part, to the temperature dependence of the dielectric constant. Furthermore, the d_{31} coefficient of piezoelectric polymers has been shown to be strongly dependent on temperature[7,8] as has the d_{31} coefficient of modified $PbTiO_3$ ceramics.[9] A detailed model of the 0-3 composite would aid in sorting out the various sources of this temperature dependence.

In conclusion, laser Doppler velocimetry has been demonstrated to be an effective technique in determining piezoelectric coefficients of materials. The effect of temperature on the piezoelectric d_{33} coefficient is largely attributed to the temperature dependence of the dielectric constant, whereas the d_{31} coefficient is observed to vary over an extended range of temperature. Furthermore, the values of d_{33}, d_{31}, and d_h are reasonably consistent in the region where the temperatures of the measurements overlap.

ACKNOWLEDGMENTS

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