Effects of High Static Stress on the Piezoelectric Properties of Transducer Materials

Piezoelectric ceramic elements in high-power acoustic transducers are subjected to high static as well as dynamic stress. This is particularly true of well-matched transducers operating in deep water, since the static stress in the piezoelectric element may be several times the water pressure. The present study was undertaken in an effort to determine the effects of static compressive stress on the piezoelectric properties of two commercial lead titanate zirconate compositions, PZT-4 and PZT-5, and of two barium-titanate compositions, commercial Ceramic B (a barium calcium titanate), and the composition 88 wt% barium titanate, 12 wt% lead titanate (BaPb12Ti). The permanent effects of stress exposure, determined at zero stress after exposure to a given stress, were found to be more severe with stress parallel to the polar axis than with perpendicular stress, as expected. Under maintained stress, however, the effects of perpendicular stress are more severe. PZT-4 and BaPb12Ti, generally better suited for use as radiating transducers, show effects dependent upon exposure time but independent of the number of stress cycles. Ceramic B and PZT-5 show effects dependent upon the number of stress cycles and less dependent upon the total period of stress exposure. Of the compositions tested, PZT-4 and BaPb12Ti were least affected by high static stress, suffering relatively little from exposure to stress as high as 15 000 psi. Of these two compositions, PZT-4 has markedly higher coupling (k_{33}-0.64 compared to 0.365) and therefore offers higher transducer bandwidth.

I. INTRODUCTION

In recent years piezoelectric ceramics have been used increasingly in underwater transducers. These ceramics are rendered piezoelectric by a poling process which consists of exposure to a high dc field. During this poling process, domains within the crystallites of the ceramic are aligned to a considerable degree, resulting in a permanent polarization. This domain alignment is accompanied by dimensional changes in the ceramic. In barium titanate the ceramic increases in length in the polar direction by 0.11% and decreases in length in all directions perpendicular to this by about 0.046%. Lead titanate zirconate compositions, with approximately twice the crystal distortion of $BaTiO_3$, typically experience strains of 0.47 and 0.20% in these directions, over four times as great as those for $BaTiO_3$ This is due to a greater degree of domain alignment during poling, which partly accounts for the stronger piezoelectric effect in the PZT compositions.

In view of the dimensional effects discussed above it is clear that properly oriented stress applied during poling can aid or impede the poling process. A tensile stress applied parallel to the field will tend to aid poling, but a compressive stress in that direction will impede the process. A two-dimensional compressive stress in the plane perpendicular to the poling field will aid poling, but if this stress is tensile, it will impede the process. A stress alone cannot pole a ceramic, since it cannot choose a parallel versus an antiparallel. arrangement of aligned domains. However, a stress alone can cause partial or complete depolarization. This fact is of considerable importance in modem underwater-transducer design, with its increasing emphasis on deepwater operation. In general, two configurations of ceramic elements have been utilized: One involves acoustic radiation in the direction of the polar axis of the ceramic, and is called the longitudinal mode; the other involves acoustic radiation in a direction perpendicular to the polar axis of the ceramic, and is called a transverse or lateral mode. In each case a solid horn is ordinarily used, at least in radiating transducers, in order to provide a better acoustic match between the transducer and water. In a typical transducer, the area ratio of the horn to the ceramic may be 10 to 1. If the transducer is immersed in water to a depth of 1000 ft the actual stress in the ceramic is 4300 psi, 10 times the hydrostatic pressure. Another transducer configuration which provides an acoustic match to the impedance of a water load is a spherical shell poled radially. In this case the major stress is a two-dimensional compressive stress perpendicular to the polar axis of the ceramic (in the plane of the shell). This stress is approximately D/4t times the hydrostatic pressure of the water, where D is the diameter and t the wall thickness of the shell.

Whether a longitudinal or a lateral mode is used, the compressive stress provided by a water load is always parallel to the direction of the ac stress amplitude which provides acoustic radiation. One would therefore expect some depoling effects for the longitudinal mode, while the lateral compressive stress required for operation in a lateral mode should have no adverse effect, because contraction takes place in this direction during poling. The picture is not this simple, however, and requires a more careful consideration of domain-switching processes. Effects of stress exposure, measured at zero stress after exposure to a given stress, are, indeed, less severe for the lateral than for the longitudinal mode (Secs. II and III), but effects measured under maintained stress are less severe for the longitudinal mode (Secs. V and VI). The high piezoelectric coupling of the longitudinal mode (k_{33} ~$2.2k_{31}$) is required for maximum bandwidth transducers.

The spherical shell, a transducer subjected to twodimensional compressive stress, provides an intermediate bandwidth ($k_p = 1.7k_{31}$). The effects of twodimensional compressive stress on the free dielectric constant were found to be quite severe by Brown.' This is due to increased domain orientation beyond that provided by the poling field, the dielectric constant being markedly reduced because of the lower dielectric constant along the polar direction in a single domain. This type of domain reorientation should, however, not lead to substantial change in the polarization and piezoelectric coupling due to the inability of a stress to distinguish between a parallel and antiparallel domain alignment.

In this study, the effects of exposure to high lateral and longitudinal compressive stress were determined for four ceramic compositions; PZT-4, PZT-5, Ceramic B, and BaPb12Ti. PZT-4 and PZT-5 are compositions based on lead titanate zirconate, while Ceramic B and BaPb12Ti are compositions based on barium titanate. PZT-4 and BaPb12Ti are best suited for high-power radiating transducers, while PZT-5 and Ceramic B are better suited for hydrophones. Table I shows typical values for some of the more important parameters of these materials. The piezoelectric coupling factors are considerably higher for the two PZT compositions than for the barium titanate compositions. In particular, the lateral coupling factor k_{31} is nearly as great for PZT-4 as the longitudinal coupling factor k_{33} for BaPb12Ti. For moderate bandwidth one then has a choice between BaPb12Ti in a longitudinal mode or PZT-4 in a lateral mode. The latter is often more convenient from a practical standpoint.

TABLE I. Elastic, piezoelectric, and dielectric properties of several ceramic compositions.

Coupling factors	PZT-4	PZT-5	Ceramic B	BaPb12Ti
k_{33}	0.64	0.675	0.490	0.365
k_p	0.52	0.54	0.325	0210
k_{31}	0.31	0.32	0.190	0.125
Piezoelectric constants				
d_{33}	270	320	150	90
d_{31}	-117	-140	-58	-30
Free dielectric constant				
K_3	1350	1500	1200	850
Elastic constants				
$1/s_{11}^E = Y_{11}^E$	8.15	6.75	11.6	12,8
$1/s_{33}^E = Y_{33}^E$	6.70	5.85	11.1	12.3
Density	7.5	7.55	5.5	5.7
Mechanical Q	500	75	400	1200
Curie point	328°C	365°C	115°C	150°C

d, 10^{-12} m/v.
1/s= Y, 10^{10}Newton/m^2
Density, 10^3 kg/m^3

All the specimens were produced by Clevite Electronic Components Division of Clevite Corporation. .Those exposed to longitudinal stress were generally 1/2-in. diamx1/8in. thick disks. Previous tests had shown very little hydrostatic stress for this diameter/ thickness ratio. Lateral tests were made on 1/4 x 1/4x1-in. bars with a pair of large faces electroded and the stress applied endto-end. Stresses were applied through hardened steel anvils and a ball-socket alignment jig using a Dillon tester with capacity for 10 000-lb force. Sample faces in contact with the steel anvils were lapped flat.

The effects of exposure to static stress, with all properties measured after stress removal, will be described first. These effects were studied as functions of the time of exposure as well as stress amplitude. Next the permanent changes due to lateral and longitudinal stress cycles will be discussed. In this case the total exposure period was very short. Then effects of longitudinal compression at elevated temperature will be described. A small oven, controlled by a Wheelco temperature controller, enclosed the anvil assembly to provide the elevated temperature. Finally, results of piezoelectric measurements under maintained static stress will be summarized. In this case the stress was applied through a compliant medium, and the piezoelectric response was determined by measurement of the strain resulting from an applied 60-cps electric field. Here specimens were slightly larger, approximately 1x1x1 in. to provide room for strain gauges for measuring the strain. A complete measurement, up to 20 000 psi and back, took about 30 min, allowing as little time as necessary to establish a fairly stable condition.

11. EFFECTS OF LATERAL AND LONGITUDINAL COMPRESSIVE STRESS WITH PROPERTIES MEASURED AFTER STRESS RELEASE

It is important to note first that exposure to stress, temperature, or electric field begins a new aging cycle in the ceramic. This is illustrated in Fig. 1, where the effects of 1-hr stress exposure on the properties of PZT-4 are shown. Two curves are given for d33, and two curves also for the dielectric constant. In each case, data for the upper curve were measured 15 min after release of
the compressive longitudinal stress, and data for the lower curve were measured three days later. A fresh

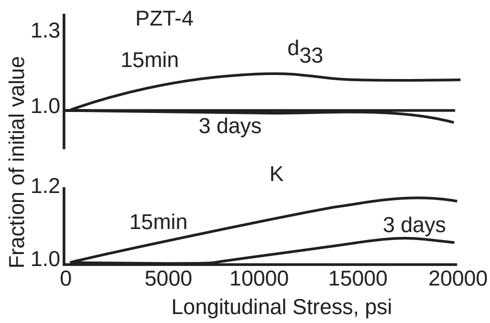

FiG. 1. Effects of exposure to longitudinal compressive stress on the properties of PZT-4. Stress applied for I hr, values measured 15 min and 3 days after stress removal.

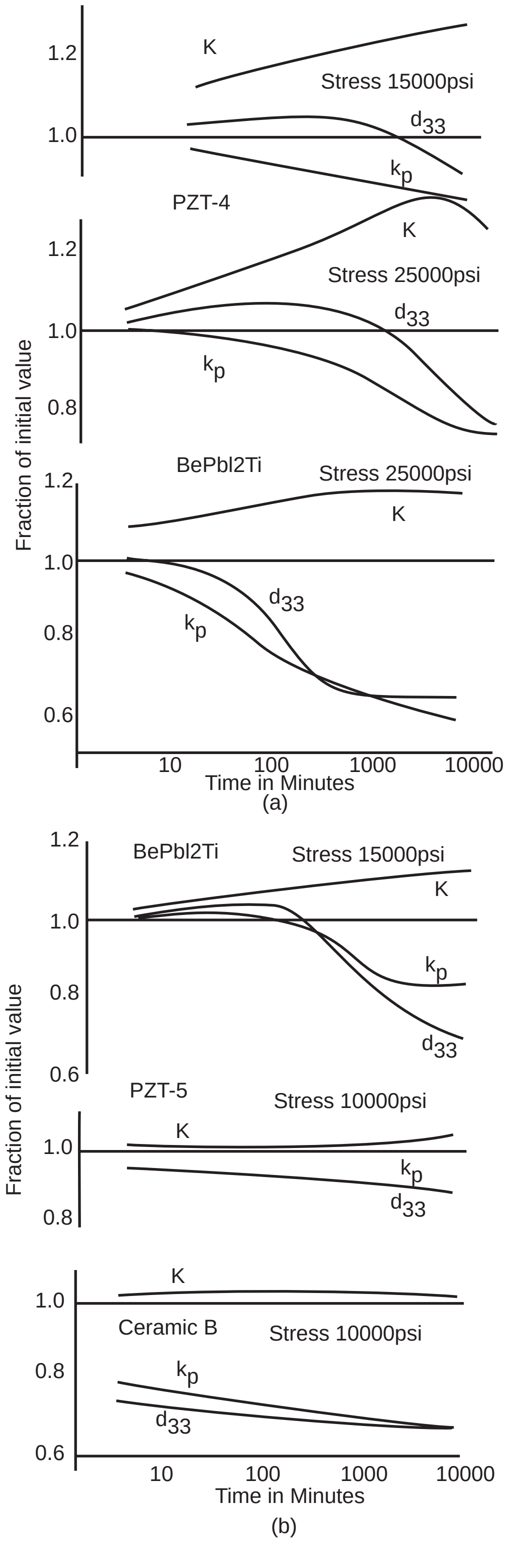

FiG. 2. Effects of exposure to longitudinal compressive stress on properties of piezoelectric ceramics.

specimen was used to obtain the data for each value of stress in order to eliminate any effects dependent upon past history.

Figures 2 and 3 show the effects of longitudinal and lateral compressive stress on d33, kp, and the dielectric constant. Each point represents data for a fresh specimen, and the measurements were made 10 min after removal of the stress. This is near the beginning of the new aging cycle, as discussed in the previous Paragraph hence, some downward aging must be expected.

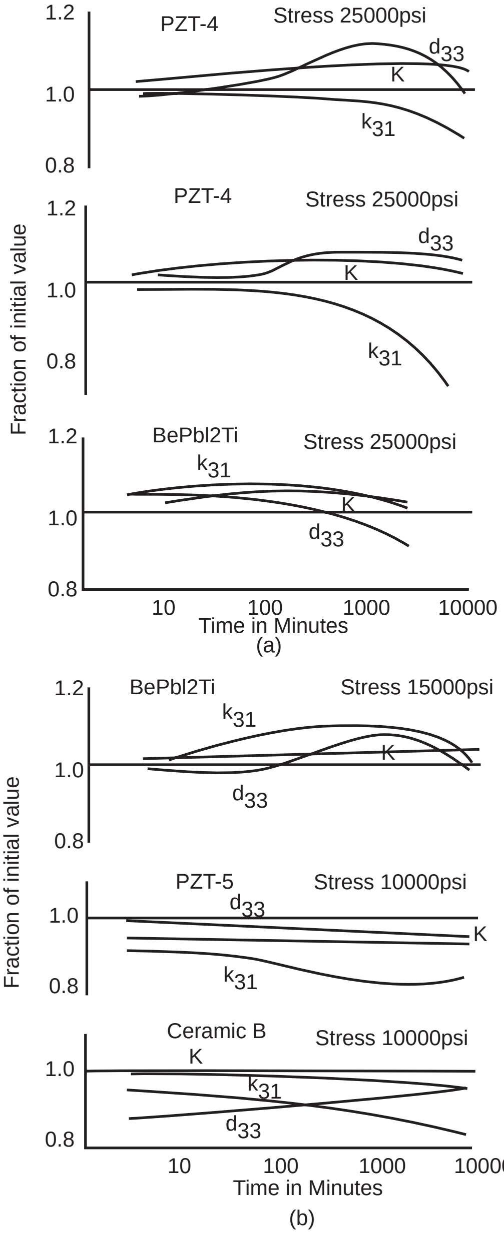

FiG. 3. Effects of exposure to lateral compressive stress on properties of piezoelectric ceramics.

stress of 10 000 psi was used on Ceramic B and PZTsince other tests showed that higher stresses cause severe degradation of piezoelectric properties, especially with the former. With PZT-5 a longitudinal stress of 10 000psi caused a few percent increase in the dielectric constant and a small decrease in d33. Exposure time had relatively little effect. Lateral stress of 10000 psi had very little effect on d33, but caused a 10% decrease in k31. There was a small decrease in dielectric constant. The opposite effects of longitudinal and lateral compression on the dielectric constant are consistent with the fact that the dielectric constant of a single domain is lower along the polar (long) direction than in any other direction. With Ceramic B results were in general similar to those obtained with PZT-5 but the reduction in piezoelectric output by parallel compression was much more pronounced (nearly 30%). Here again there was relatively little dependence on time of exposure.

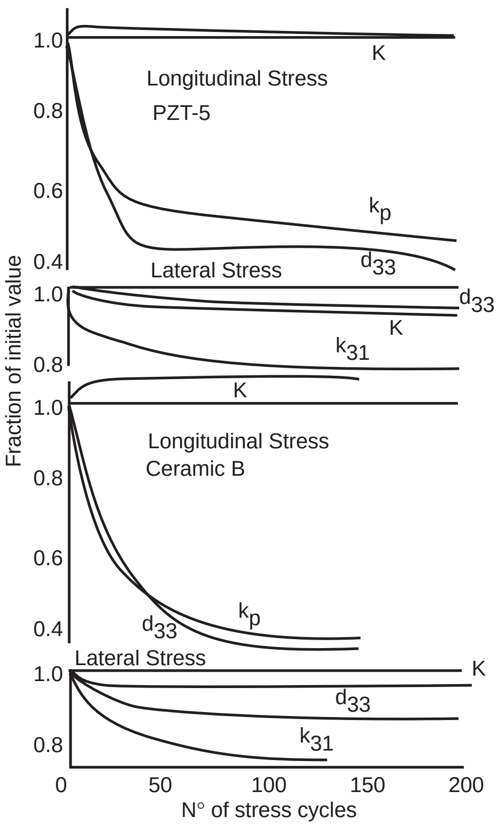

FIG. 4. Effects of longitudinal and lateral stress cycles to 10 000 psi on the properties of PTZ-5 & Ceramic B.

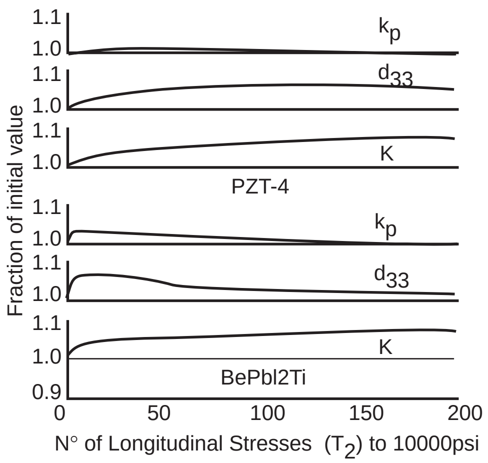

FIG. 5. Effects of longitudinal stress cycles to 15 000 psi on the properties of PZT-4 and BaPb12Ti.

The effects of exposure to longitudinal compression for PZT-4 and BaPb12Ti are also shown in Figs. 2 and 3. The effects are more severe for the barium titanate composition, but in both cases the effects are dependent upon the length of the exposure period. In general the effects of lateral stress are almost as severe as the effects of longitudinal stress. It will be noted that the main effect of lateral stress exposure is a decrease in the lateral response k_{31}, and that there is relatively little effect on the longitudinal response d_{33}. In any case where the compressive stress in a water-loaded transducer is directed along a lateral direction, it is this direction which is relevant; its effect on k_{33} is not important.

III. EFFECTS OF LATERAL AND LONGITUDINAL COMPRESSIVE STRESS CYCLES

In Sec. II it was shown that the permanent effects of stress exposure, determined at zero stress after exposure to a given compressive stress, are relatively independent of exposure time for PZT-5 and Ceramic B but not for BaPbl2Ti and PZT-4 When the effects of stress cycles

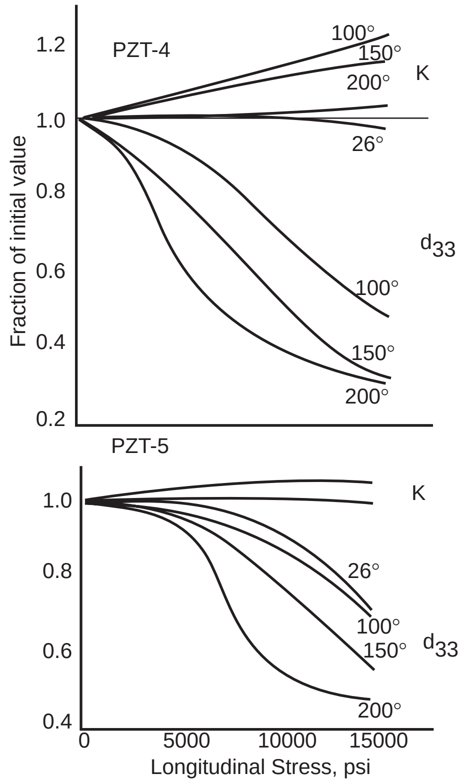

FIG. 6. Effects of exposure to longitudinal compressive stress onproperties of PZT-4 and PZT-5 at elevated temperatures.

were determined, it was also found that PZT-4 and BaPbl2Ti exhibit behavior opposite to PZT-5 and Ceramic B. Here the total exposure time was limited to about 15 sec/cycle in order to separate the effects of exposure time and of the stress cycle itself.

Even though PZT-5 and Ceramic B suffer changes fairly independent of the time of exposure to 10 000 psi lateral or longitudinal compressive stress, changes due to stress cycles to 10 000 psi are very severe. Data are shown in Fig. 4. With PZT-5, k_p and d_{33} decreased linearly as functions of the number of stress cycles to about 65% of their original values for the first 20 longitudinal stress cycles, leveling off near 50% and 40%, respectively. With lateral stress k_{31} was reduced to 80% of its original value, but d_{33} was relatively unaffected. Roughly similar but somewhat more pronounced changes were obtained with Ceramic B.

For both Ceramic B and PZT-5 the effects of stress cycles were markedly more pronounced than those of much longer total exposure time to the same stress. These compositions might be suitable for use in deepwater hydrophones which are permanently anchored, but neither should be subjected to pressure cycles.

Figure 5 shows similar data for PZT-4 and BaPb12Ti. Here there was very little dependence on the number of stress cycles, even though pronounced changes occurred when the same stresses were applied for a period of time (see Figs. 2 and 3).

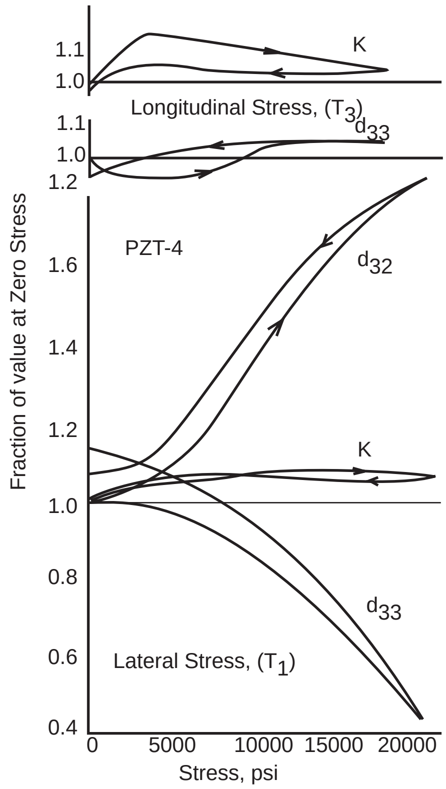

FIG. 7. Effects of longitudinal and lateral compressive stress on the properties of PZT4; measured with stress maintained.

IV. **EFFECTS OF LONGITUDINAL COMPRESSION AT ELEVATED TEMPERATURES**

The effects of exposure to longitudinal compression at 100, 150, and 200°C are shown in Fig. 6 for PZT-4 and PZT-5. Here again a fresh specimen was used to obtain data for each point. The degradation of properties at the higher temperatures was less severe with PZT-5, but that due to stress cycling as previously discussed would alter this picture in favor of PZT-4. Similar data are not available for the barium titanate compositions, but a few tests made at 70°C indicate that effects are more severe than those suffered by PZT-4 at 100°C

V. **PIEZOELECTRIC AND DIELECTRIC PROPERTIES MEASURED UNDER MAINTAINED COMPRESSIVE STRESS**

All data presented up to this point were obtained at zero static stress after a specified exposure to lateral or longitudinal compressive stress. It was noted that the permanent effects of exposure to lateral compression are somewhat less severe than those resulting from exposure to longitudinal compression. When data are measured while the static stress is maintained, very different results are obtained.

Figures 7 and 8 show the dielectric constant and d-constants as functions of maintained static longitudinal and lateral compressive stress Jor PZT-4 and

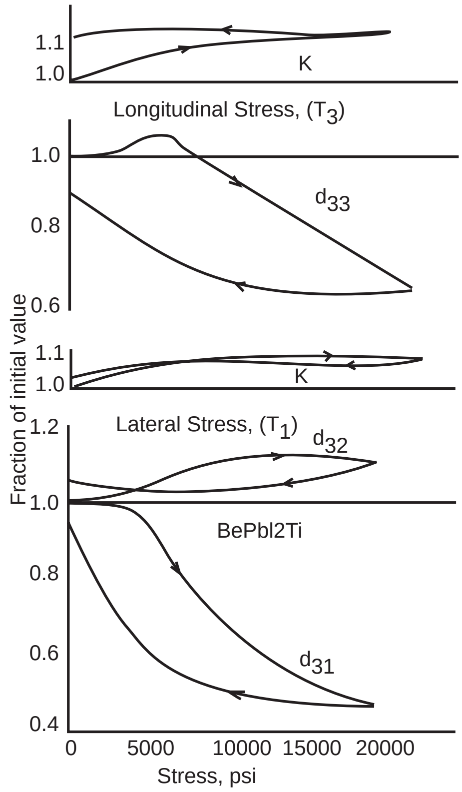

FIG. 8. Effects of longitudinal and lateral compressive stress on the properties of BaPbl2Ti; measured with stress maintained.

BaPbl2Ti. With PZT-4 the changes due to longitudinal compression were quite negligible up to 19 000 psi, the limit of this test. BaPbl2Ti suffered a decrease of nearly 40% in d_{33}, but the change in capacitance was moderate. Under lateral compression the normal isotropy in the plane perpendicular to the polar axis was disturbed for both compositions, and therefore d_{31} or d_{32}. This inequality remained even after the stress was removed, but it was then much less pronounced. The effect of the stress is to reduce drastically the piezoelectric response d_{31} in the direction of compression and to increase drastically the response d_{32} in the irrelevant (unloaded) direction perpendicular to the plane containing the polar axis and the axis of lateral compression. Comparison of Figs. 2 and 3 with Figs. 7 and 8 leads one to the conclusion that, even though the permanent degradation due to exposure to lateral compression is somewhat less severe than that due to exposure to longitudinal compression, changes under lateral bias stress are much more pronounced than those under longitudinal bias stress.

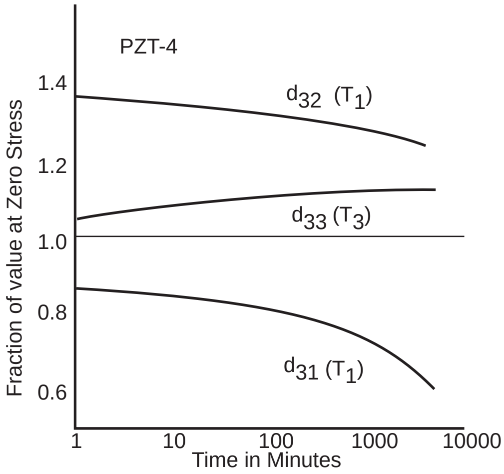

FiG. 9. Aging of d31 and d32 at 10 000 psi lateral stress; aging of d33 at 10 000 psi longitudinal stress; all measured with stress maintained; PZT-4.

VI. **AGING UNDER MAINTAINED COMPRESSIVE STRESS**

In view of the data of Fig. 1 one might expect severe aging under maintained static stress. Figure 9 shows that aging under 10 000 psi longitudinal stress is considerably less severe than that under 10 000 psi lateral stress for PZT-4 Furthermore, aging effects under maintained longitudinal compression are less severe than those encountered after removal of the stress.