Applied Acoustics **41** (1994) 299–324

Materials for High Temperature Acoustic and Vibration Sensors: A Review

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ABSTRACT

The industrial and scientific communities have expressed a real need for the capability of pressure, acoustic, and vibration sensing at elevated temperatures. This review compares the various commercial methods and materials for acoustic transduction, identifying their advantages and limitations. Techniques and devices include simple piezoelectric sensors, accelerometers, strain gauges, proximity sensors, fiber optics and buffer rods. Sensors with operating temperatures in excess of 650°C are readily available from commercial sources. Of the mechanisms investigated, the piezoelectric approach offers several advantages, including design cost and simplicity. Therefore, the bulk of this review concentrates on piezoelectric materials, both those that are already available commercially, and those that are presently under development. The new materials include perovskite layer structure ferroelectric ceramics, which possess the highest known Curie temperatures, and thin film AlN, which has been reported to be piezoactive at 1150°C.

1 INTRODUCTION

High-temperature electronics is an area of research offering interesting materials and design challenges and one of significant industrial importance. The major impetus for the development of high-temperature electronic materials, devices, circuits, and systems can be credited to the energy crisis of 1974, when a commitment was made to the development of national energy resources by geothermal exploration.[1] At that time, geothermal and oil-well logging industries voiced their need for sensors and electronic

Applied Acoustics 0003-682X/94/$07.00

systems with higher operational temperatures for deep drilling in the earth's crust. The economic importance of world energy independence and reduced waste provided additional incentive for their development.

The aerospace and aircraft industries have especially difficult high-temperature requirements. With space and weight at a premium, engine designers and builders find it difficult to protect sensitive electronic systems in a cool, remote place. Electronic controls are to be placed directly inside jet engines because of reliability and noise requirements, so sensors need to be built that can withstand temperatures of 500–1000°C while allowing mission lifetimes up to 100 000 h.

In automotive electronics, the number of sensors and actuators continues to increase each year. Ceramic and semiconductor sensors which record temperature, oxygen pressure, and preignition knock are used in conjunction with microprocessor-based controls to improve the efficiency and reliability of internal combustion engines.[2] Further efficiency can be realized by operating combustion engines at higher temperatures. Research on the use of ceramic components in a diesel engine has led to higher operating temperatures, resulting in a potential increase in fuel efficiency up to 65%, along with a notable reduction in exhaust pollution.[3] Higher operating temperatures do, however, place additional requirements on the sensors. Environments of 150°C with repeated temperature cycles are at present considered the automotive norm, and higher temperatures are expected in the future.

Until the present, there has not been a review of the types of sensors commercially available for high-temperature acoustic sensing applications. The purpose of this paper is to provide such a review. Included in the discussion are the commercially available high-temperature piezoelectric materials used in many of the sensors described. In addition, new advances in materials that may permit the design of sensors which can operate at temperatures well above those currently available will be presented.

2 ACOUSTIC MEASUREMENT TECHNIQUES AND APPLICATIONS

Critical parameters for acoustic and vibration sensors involve the measurement of dynamic pressure pulses, vibrations (relative and absolute), acoustic emissions, strains, and the dynamic proximity of machine components. An example of a dynamic pulse is the rapid increase in pressure upon ignition of the air–fuel mixture in the cylinders of an internal combustion engine. The timing and the shape of the pulse have a large effect upon engine efficiency. Transducers fitted into the cylinder head have been used for monitoring combustion pressure to optimize ignition timing.[4]

Dynamic monitoring is also employed in such machines as aircraft engines, gas turbines, and power generators, all of which have high speed rotors. Sensors strategically mounted on the machine detect destructive conditions of imbalance, or unequal loading of the rotor, enabling the possibility of corrective measures to be implemented. The sensors convert the associated mechanical energy of the vibration to electrical energy which can then be amplified and monitored. Computer coupling, allows real-time status reports of the condition of the machine while providing a comparison with an operating norm.

Non-destructive testing (NDT) is another widely used application of acoustic sensors that involves either passive sensing of 'acoustic emissions', or active 'ultrasonic testing' techniques. An application of the first method involves affixing a number of acoustic sensors in strategic locations on the wall of a vessel, such as a chemical storage tank, pressurizing the vessel to create strains, and examining the acoustic emissions which result from the Kaiser effect.[5] Cracks and poor joints can then be detected and logged so that repairs can be made. This method has also been successfully applied in the inspection of the fuselage of large aircraft to detect cracks resulting from fatigue. The ultrasonic method of NDT incorporates a transducer to generate an acoustic signal at ultrasonic frequencies that is transmitted through the test specimen. When the acoustic wave reaches an interface of the sample it is reflected back to the transmitter/sensor which, therefore, acts as a transceiver. If, however, the wave impinges upon a flaw, a portion of the wave is reflected and thus reaches the sensor ahead of the original wave. This, then, becomes a valuable tool for locating defects within a structure.[6] Figure 1 is a simple illustration of the technique. Acoustic sensors also find use in the hostile environments of deep oil wells for seismic data logging, and in nuclear power plants to monitor the condition of heat exchange pumps and pipes.[7]

An indirect way of sensing vibration is by measuring capacitive changes in an air gap. An air gap sensor placed in close proximity to a

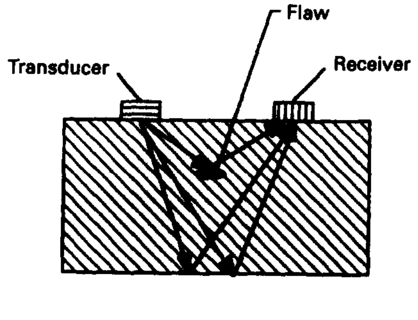

Fig. 1. Detection of a flaw in a solid by ultrasonic testing (NDT).

TABLE 1
Acoustic and Vibration Measurement: Industrial Applications

Aerospace	Measure rotational speed of jet turbine rotors
	Space vehicle acceleration
	Trajectory monitor
	In-flight vibration monitor
	Nozzle pressure
	NDT test of air frames
Automotive	Knock sensor
	Fluid level sensor
	Crash test
	Vibration control
Industrial	Dynamic pressure monitors
	Vibration detectors
	Flow detectors
	NDT testing
	Proximity detectors
	Noise 'fingerprinting'
Medical	Diagnostic imaging
	Impact sensors
Military	Hydrophones
	Range finders
	Noise detection in vehicles
	Security systems
Power generation	Leak detector
	Liquid sodium coolant pump monitor
	Nuclear reactor monitor
	Air gap monitoring between stator and rotor
	Fuel rod monitor
	Coal feeder monitor
	NDT of pressure vessels
Commercial	Fish finders
	Phonographs pick-up
	Microphones

high-speed rotor will detect dynamic changes in the spacing between the two which, in effect, is an indicator of an imbalance condition or some other malfunction. This type of sensor has found wide acceptance in hydroelectric power plants.[8]

Examples of pressure. acoustic and vibration measurement techniques described above serve to illustrate the wide variety of possible applications. Additional applications are listed by industry in Table 1. These are routinely employed in the range of $-55°C$ to $125°C$, however, several devices are needed to operate in much harsher thermal conditions. Sensor designs and their respective temperature limitations are described in the following section.

3 SENSOR DESIGNS

Listed in Table 2 are current commercial sensor designs. They include piezoelectric discs or plates, accelerometers, strain gauges, proximity sensors, fiber-optic sensors, and systems incorporating buffer rod extensions. Brief descriptions of these devices are as follows.

3.1 Simple disc

The most basic sensor incorporates a simple disc, or other simple shape, composed of a piezoelectric ceramic, which is attached directly to the wall or frame of an engineering structure, embedded in a recess, or mounted in a replaceable fixture. Figure 2 is a schematic construction of a piezoelectric knock sensor using a bending mode resonance to detect vibrations. The piezoelectric element 'generates' a voltage in response to stresses caused by the acoustic energy impinging upon it.[9] The magnitude of the voltage generated is directly related to the product of the applied stress and the piezoelectric voltage or 'g' constant of the material. The electrical signal is then amplified and fed to a microprocessor in the control system. This arrangement is capable of extremely high sensitivity on the order of pico (10^{-12}) strains. It is also self-generating, rugged, low-cost, and simple. The temperature limitations of these sensors arise from the loss of piezoelectric properties that occur as the material approaches its transition (Curie) temperature T_c, a topic which will be discussed later. For lead zirconate–titanate, (commonly called PZT), the most widely used piezoelectric ceramic material, the maximum use temperature is ~200°C. Other temperature limitations result from the failure of the adhesives used to mount the disk, and melting of the solder used to attach the leads.

3.2 Buffer rod extensions

For many high-temperature applications, buffer rod extensions are utilized to transmit and receive acoustic signals from very hot areas, effectively

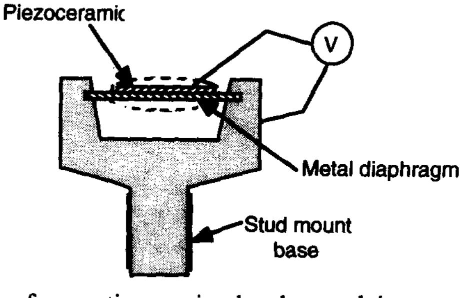

Fig. 2. Two methods of mounting a piezobender used in automotive knock sensors (after Ref. 9).

TABLE 2
Comparison of Commercial Strain, Vibration, and Acoustic Senors

Operation principle	*Piezoelectric disc or plate*	*Accelerometer*		*Strain gauge*	*Proximity sensor*		*Buffer rod*	*Fiber-optic sensor*
	Piezoelectric	*Piezoelectric*	*Magnetic induction*	*Resistive metal*	*Capacitive*	*Eddy current*	*Piezoelectric*	*Laser/optic*
Measured parameter	Acoustic waves vibration	Acceleration absolute dynamic	Vibration pressure	Load, force vibration absolute	Relative air gap displacement	Vibration	Acoustic waves	Strain vibration
Present maximum temperature (°C)	400	650	480	850	150	450	1 100	300
Temperature stability	Good	Good	Good	Fair	Good	Fair	Good	Poor
Temperature sensitivity	Good	Good	Good	Fair	Good	Fair	Good	Poor
Resolution/ sensitivity	High	High	Fair	Low	Low	Low	Fair	High
Frequency range	0·05–1 MHz	0·05– MHz	0·015–2 kHz	DC–kHz	DC–kHz	DC–10 kHz	0·05 kHz–MHz	DC–10 kHz
Power consumption	Low	Low	Low	Medium	Medium	Medium	Low	High
Cost and complexity	Low	Medium	Medium	Low	Low	High	High	High
Durability	Good	Good	Good	Fair	Good	Fair	Good	Poor

isolating temperature-sensitive transducers from hostile environments. The rod serves as an acoustic wave guide to couple the hot test specimen with the sensitive piezoelectric transducer. The rods are generally constructed of stainless-steel and are cooled with water or air to prevent damage to the transducers. This arrangement permits the use of a conventional piezoelectric material, which would otherwise lose its piezoelectric properties at high temperatures. This technique is widely used on NDT probes in the metals industry to detect cracks and other defect in hot steel blooms and pipes at temperatures in the neighborhood of 1100°C.[10] A similar application employs hollow 'buffer pipes' to couple dynamic pressure sensors with jet and rocket engines.

3.3 Accelerometers

Accelerometers most often use a piezoelectric as the internal sensing element. The various designs exploit different mechanisms to translate mechanical energy to a measurable response, but all operate based on Newton's second law:

$$F = ma$$

The accelerometer shown in Fig. 3 differs from the simple disk piezoelectric sensor in that a seismic mass (m) is attached to the piezoelectric element and the assembly is hermetically sealed in a protective case.[11] As a response to acceleration (a), the mass imparts a force (F) on the element, which in turn generates a voltage in proportion to the magnitude of the stress. Depending upon the sensitivity and temperature range required for the application, transducer manufacturers utilize several different piezoelectric materials. Commercial accelerometers using lithium niobate ($LiNbO_3$) single crystal elements are rated for continuous use up to 650°C.[12,13]

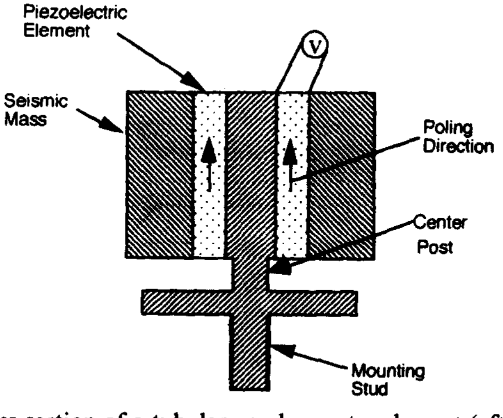

Fig. 3. Cross-section of a tubular accelerometer element (after Ref. 11).

Accelerometers based on magnetic induction, employ a permanent magnet as the seismic mass and a field coil for the active element. The mass is partially restrained by a spring, but when the mass is accelerated, it moves relative to the coil, thus inducing an electromotive force. As with the case of piezoelectric types, the resulting electric signal must be amplified and filtered before it is fed to the instrumentation. The upper temperature limit of this type of device is determined by the Curie temperature of the permanent magnet. Commercial units are rated for temperatures as high as 480°C.[14]

3.4 Strain gauges

Figure 4(a) is an illustration of a bonded metallic foil strain gauge.[15] Resistive strain gauges involve a change in electrical resistance (ΔR) resulting from the mechanical strain ($\Delta L/L$) of the sample to which the gauge is bonded. The sensitivity is determined by the gauge factor (GF):

$$\mathrm{GF} = \frac{\Delta R/R}{\Delta L/L}$$

With a typical gauge factor of 2, and a nominal resistance $R = 100\ \Omega$, these gauges require an ohmmeter with very high sensitivity to measure the ΔR accompanying a strain of 1×10^{-6}. A more accurate way of measuring small changes in resistance incorporates a Wheatstone bridge as shown in Fig. 4(b).

To compensate for the nonlinear character of the thermal coefficients of expansion of the alloys used, an unstrained reference gauge is installed in an adjacent bridge arm. This second gauge is subjected to the same temperature as the first, thereby effecting electrical cancellation of the apparent strain. Commercial metal gauges are typically limited in temperature range to about 300°C; however PtW wire gauges are available for

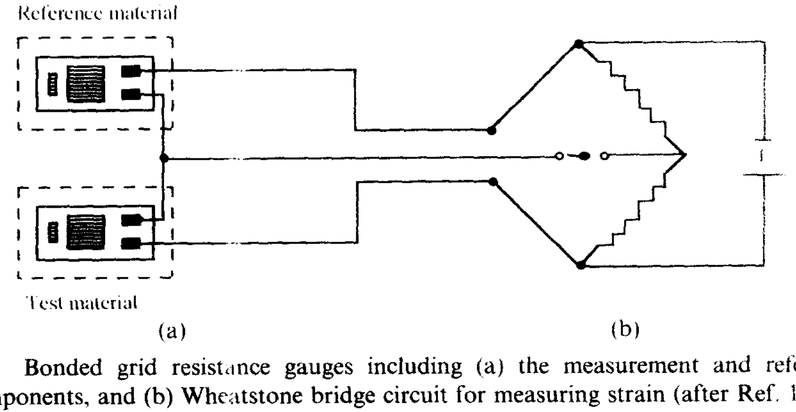

Fig. 4. Bonded grid resistance gauges including (a) the measurement and reference components, and (b) Wheatstone bridge circuit for measuring strain (after Ref. 15).

use up to 850°C,[16] and other exotic metals such as PdCr are being studied for use up to 1000°C.[17]

Metal strain gauges are simple in design, low-cost, and can detect static strains. Even using the Wheatstone bridge configuration, however, strain sensitivity is low compared with piezoelectric transducers.

3.5 Proximity sensors

Proximity sensors are often used to monitor the degree of rotation of the armatures of electric generators as well as the clearance between the armature and the stator. This serves as an indirect method of measuring vibration if an unbalanced condition exists. The two types of proximity sensors available commercially are (i) air gap capacitors, and (ii) eddy current detectors. Both types require electronic conditioners to supply an input signal, and processors to analyze the signals modified by the sensors.

(i) An example of an air gap capacitor is illustrated in Fig. 5.[18] In this application on an electric power generator, the armature serves as one plate of the capacitor while the other plate is an integral part of the sensor and air is the dielectric. The plates are maintained at a close proximity with one another so that a small change in spacing results in significant changes in capacitance. The change in capacitance can then be detected by the control circuitry and acted upon accordingly. The detection of variations in air gap as small as one micron are possible in installations on hydroelectric power generators. At present, commercial units are constructed of fibreglass composites for use at temperatures up to 150°C,[12] but it seems reasonable that units could be constructed from more refractory materials for higher temperature applications.

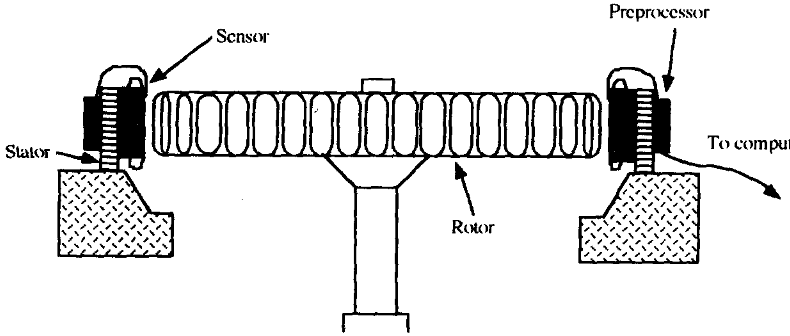

Fig. 5. Air gap sensors mounted on an AC generator (after Ref. 18).

(ii) The second kind of proximity sensor is the eddy current type. It determines the relative position of a rotating shaft by detecting changes in magnetic field between the sensor and the target shaft. The eddy current sensor operates in a manner similar to that of an electrical transformer. The sensing coil resembles the primary winding, and the small eddy current loops induced in the target by the variable magnetic field of the sensing coil act like the secondary windings. The small eddy currents cause a change in phase and amplitude of the sensing coil signal. The target material must therefore be metallic. The sensing coil is mounted near the rotating shaft or moving component of a machine with lead cables attached to the exterior end. Commercial models are currently rated for use up to 450°C.[14] The temperature limitation is primarily due to the temperature coefficient of resistance of the wire used to wind the probe coil.[19]

3.6 Fiber optic sensor interferometers

Fiber optic sensor (FOS) interferometers or phase sensors are used to detect small strains, (in the order of 10^{-7}) or vibrations in a sample without making physical contact. This capability allows measurements to be made inside a hostile environment, such as a furnace. A simplified schematic of a system is shown in Fig. 6. There are several different designs, but all are based on measuring a phase change in the transmitted light signal resulting from a change in length of the optical fiber. Optical fibers are typically SiO_2 glass; however, fibers of more heat-resistant materials such as sapphire (Al_2O_3) are commercially available. Along with the fiber, a large number of additional components, such as a laser source, phase demodulator, filters, and microprocessors, etc., are needed for these systems. High cost, complexity and sensitivity to variations in temperature are disadvantages of this approach.

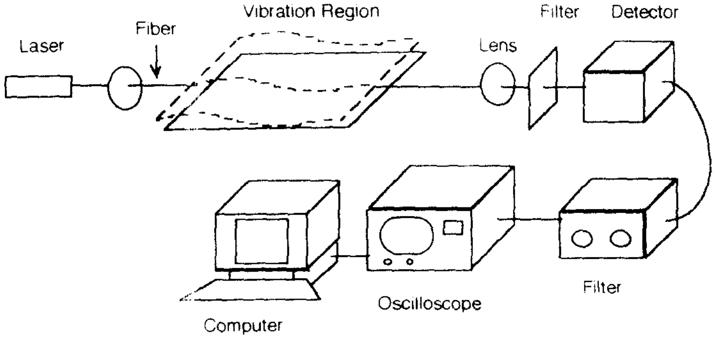

Fig. 6. Fiber-optic vibration sensor and associated electronics.

In summary, with respect to the operational parameters listed in Table 2, piezoelectric trandusers offer many advantages over the other types of acoustic sensors. Specifically, their compliment of sensitivity, stability over a wide temperature range, and simple design offers the lowest cost approach. Polycrystalline ceramics are often the material of choice because they can be readily manufactured into a variety of configurations. Therefore, the remainder of this paper concentrates on high-temperature piezoelectric materials, with a review of those currently available, and a discussion of future prospects.

4 PIEZOELECTRIC CERAMICS

4.1 Piezoelectricity

The phenomenon of piezoelectricity was first reported in 1880 by Pierre and Jacques Curie. Since that time, a number of crystalline materials have been found to exhibit piezoelectric activity, although only a few are of practical concern. The common feature of all piezoelectric crystals is that they have no center of symmetry along the piezoelectric axes. Ferroelectric ceramics possess a multitude of individual dipole domains which are distributed throughout the body in various crystallographic orientations, resulting, in a net dipole moment of zero. In a process, called poling, a relatively large electric field is applied through the body causing a common alignment of a large portion of the dipoles, thus rendering the material piezoelectric. These crystals develop an electric field when subjected to an applied stress (sensor), or conversely, exhibit a mechanical deformation with the application of an electric field (actuator). Since the details of piezoelectricity are covered in a number of excellent texts and review articles, the reader is advised to consult them for further information.[11,20–22]

4.2 Electromechanical properties

In order to describe the electromechanical properties of piezoelectric materials, a large number of interrelated coefficients are used, many of which have been standardized by the IEEE.[23] However, with regards to high-temperature materials, only the most important coefficients and properties will be presented in this section. (Note: where subscripts are used with coefficients, the first number refers to the axis of polarization, and the second refers to the axis of applied stress, or applied field.)

4.2.1 Ferroelectric Curie temperature

Most of the piezoelectric materials described in this paper are ferroelectric and capable of being polarized, with the polarization resulting from the asymmetry of its crystal structure. When the crystal is heated, its internal kinetic energy increases. At a certain temperature, called the Curie temperature (T_c), the crystal changes to a structure of a higher symmetry, the alignment of the dipoles is lost and all piezoelectric activity disappears. Upon cooling, the dipoles do not realign unless they are subjected to a strong electric field. Other consequences of increasing temperature are changes in the values of electromechanical coefficients, which become more pronounced as the T_c is neared. This can be particularly important in applications where the electrical properties of the sensor are closely matched to the instrumentation. In addition, dipoles that were polar oriented have a tendency to reverse back to their original position thus degrading the piezoelectric effect in a process known as 'thermally activated aging'. Generally, a maximum operating temperature of one-half the T_c is considered safe.

4.2.2 Resistivity and the RC time constant

High electrical resistivity is necessary so that a large field can be applied during poling without breakdown or excessive charge leakage. High insulation resistance '*R*' is also required during operation of the device. The transducer must not only develop a charge for an applied stress or strain, but must also maintain the charge for a time long enough to be detected by the electronic system. The length of time the charge is maintained is proportional to the *RC* time constant (resistance × capacitance). The minimum useful frequency of a sensor, known as the lower limiting frequency (f_{LL}), is inversely proportional to the time constant:

$$f_{LL} = \frac{1}{2\pi RC}$$

where C is the device capacitance.

Below f_{LL}, the charge will drain off before it is detected because of conduction in the sensor. With low f_{LL} the dynamic bandwidth can be extended to sonic frequencies, and thus, a large *RC* constant is desirable for many applications.

4.2.3 Piezoelectric constants

The piezo strain '*d*' constant gives the ratio of the strain developed in the specimen to the electric field applied at the electrodes, and conversely, the ratio of short circuit charge, per electrode area, to the applied stress. The stress can be applied to the body in different modes as illustrated in

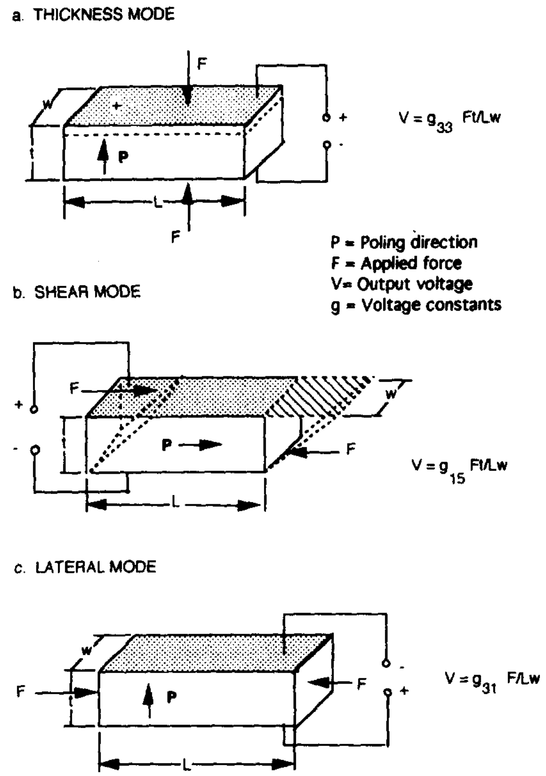

Fig. 7. (a) Thickness, (b) shear, and (c) lateral mode piezoelectric sensors. P indicates the poling direction. The output voltage (V) is proportional to the applied force (F) and piezoelectric voltage coefficient (g_{ij}) of the material.

Fig. 7. The piezoelectric voltage 'g' constant gives the ratio of the field developed to the stress applied, and conversely, the ratio of strain to the applied charge to the electrode area. The 'g' constant is related to the 'd' constant by the permittivity, $K\varepsilon_0$.

$$g = \frac{d}{K\varepsilon_0}$$

In comparing the properties of high-temperature piezoelectric materials, the 'g' constant tends to be the more meaningful coefficient.

4.2.4 *Mechanical* Q

The mechanical Q expresses the ratio of strain in phase with stress to strain out of phase with stress in the vibrating element, or in other words, the relative amount of input electrical energy that is converted to mechanical energy as opposed to that which is lost as heat. Piezoelectric materials with high 'Q_m' factors are characterized by having narrow resonant peaks, whereas, those with low 'Q_m' factors have broader bandwidths.

4.2.5 Sensitivity
For a given stress applied to the device, the output voltage generated by the piezoelectric 'generator' should be sufficiently high to be detected above background noise. The most sensitive devices produce the greatest output per unit of stress. Unfortunately, for high-temperature sensors, ferroelectric materials exhibit a considerable increase in permittivity, with increasing temperature, a condition which effectively reduces the voltage constant 'g'. Every piezoelectric material reaches some temperature at which its voltage constant 'g' and resistivity are diminished to the point where the output becomes undetectable. In practice, the relatively small output signals generated by high temperature accelerometers necessitate the use of electronic charge amplifiers for enhancement, but there is a practical limit to how small a signal that can be accurately detected.

5 COMMERCIAL PIEZOELECTRIC CERAMICS

Table 3 lists typical piezoelectric ceramic materials found in commercial accelerometers and vibration sensors, along with reported room temperature properties.[21,24–28] Owing to the lack of reliable published high-temperature data available for this work, samples were obtained, their electrical properties were measured at elevated temperatures, and the data reported in Table 4.[25–29] In addition, the resistivity of each material was measured over a wide range of temperatures as shown in Fig. 8.[25–28,30–34] Good agreement was found with values reported in the literature (as shown in Fig. 8).

A brief description of each of these piezoelectric materials, or related compositional families, is presented in this section. With the exception of quartz, all are ferroelectric.

5.1 Quartz

Quartz (SiO_2) is one of the earliest piezoelectric materials used in electronic devices. Originally, natural quartz crystals were used, but now have been widely replaced by hydrothermally-grown synthetic quartz. Because of its low mechanical loss (high Q_m), narrow bandwidth, and highly temperature-stable resonant frequency, quartz is the material of choice for timing standards and monolithic filters in communication equipment. However, as seen in Table 3, the piezo 'd' coefficient is relatively small and thus

TABLE 3

Reported Room Temperature Electrical Properties of Commercial High Temperature Piezoelectric Materials

Material	Structure	Curie point T_c (°C)	Dielectric constant K (10^{-12} C/N)	Piezoelectric strain constant (10^{-3} Vm/N)		Piezoelectric voltage constant		Electromechanical coupling (10^{12} Ω-cm)		Mechanical quality Q_m	Resistivity 20°C ρ	Ref.
				d_{33}	d_{15}	g_{33}	g_{15}	k_{33}	k_{15}			
$Pb(Zr, Ti)O_3$ (Soft PZT)	Perovskite	330	1 800	417	710	25	41	0·73	0·77	75	100	N-21[26]
$(BaPb)Nb_2O_6$ (BPN)	Tungsten bronze	400	300	85	100	32	46	0·30		15	1	K-81[25]
$PbTiO_3$ (PT)	Perovskite	470	190	56	68	33	32	0·45		1 300	10	LTT-3[28]
$Na_{0.5}Bi_{4.5}Ti_4O_{15}$ (NBT)	Bismuth	~600	140	18		15		0·15		100	1 000	K-15[25]
$LiNbO_3$ (LN)	Corundum	1 150	25	6	69	23	91	0·23	0·60	NR	1	[27]
SiO_2	α-Quartz	573	4·5	2(d_{11})		50		NR		10^5	1 000	[21,24]

TABLE 4
Selected High Temperature Properties of Piezoelectrics

Material[a]	T_c (°C)	*Vibration mode (ij)*	g (RT)[b] ($\times 10^{-3}$ Vm/N)	*TCF* (ppm/°C)	ρ (Ω-cm)	ε_r (10 kHz)	tan δ (10 kHz)	*RC* (s)	*Ref.*
PZT (DOD II) (200°C)	360	[33]	15 (25)	+200	$10^{[illegible]}$	5 000		250	N-21[26]
(BaPb)Nb_2O_6 (300°C)	400	[33]	24 (30)	−135	10^7	530	0·036	0·000 5	K-81[25]
$PbTiO_3$ (400°C)	450	[15]	21 (29)	−97	2×10^5	1 000	0·35	0·000 02	LTT-3[28]
$Na_{0.5}Bi_{4.5}Ti_4O_{15}$ (400°C)	600	[33]	10 (18)	−118	3×10^8	262	0·10	0·007	K-15[25]
$LiNbO_3$ (400°C)	1 150	[15]	93 (99)	−76	3×10^7	100	0·001	0·000 3	[27]
Sr_2NbTaO_7 (400°C)	823	[24]	5 (6)	−74	4×10^9	40	0·10	0·01	[29]

[a] PZT and BPN were evaluated at approximate maximum use temperatures, the others at 400°C.
[b] Room temperature values (RT) also shown for the voltage constant (g).

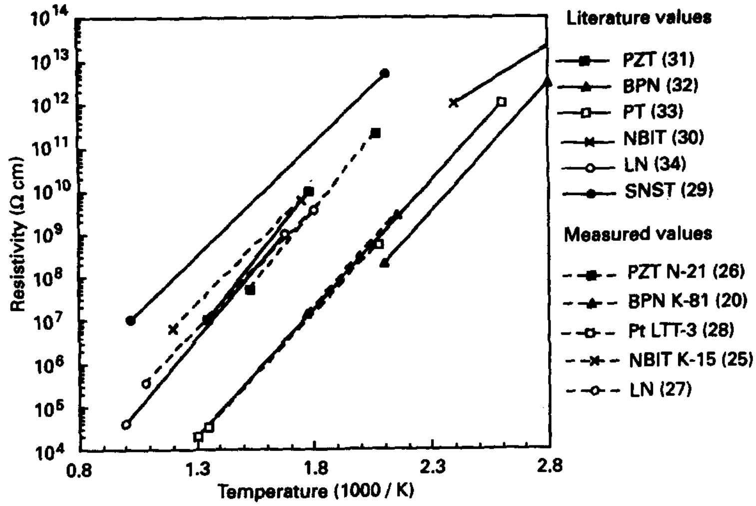

Fig. 8. Temperature dependence of resistivity for piezoelectric ceramics: 'soft' lead zirconate titanate (PZT), barium-doped lead metaniobate (BPN), sodium bismuth titanate (NBIT), lead titanate (PT), lithium niobate (LN), and perovskite layer structure strontium niobate tantalate (SNST). Solid lines represent data taken from literature, while dashed lines represent data measured from commercial materials for this work. All are polycrystalline ceramics except single crystal LN.

the amount of electric charge that can be generated is low. Although piezoelectric α-quartz has a transition temperature of 573°C, its use temperature is normally limited to 350°C. Above that temperature the crystal structure is subject to twinning, destroying its piezoelectric properties.[24]

5.2 Lead zirconate titanate

Lead zirconate titanate ($Pb(ZrTi)O_3$)(PZT) has the perovskite structure and is a solid solution of tetragonal $PbTiO_3$(PT) and orthorhombic $PbZrO_3$(PZ). Because of the large number of polarization directions available, compositions near the morphotropic phase boundary at approximately 53:47 PZ to PT are easily poled to high remanent polarization and exhibit extremely high values of electromechanical coupling coefficients and electrical permittivity. Because of its superior piezoelectric properties and higher operating temperature, PZT has largely replaced barium titanate (BT)—T_c ~ 130°C, the first widely used ferroelectric, in all but the lowest cost commercial products. Useful variations in properties of PZT can be obtained by compositional additives. Niobium-doped PZT (DOD Type II) is used extensively in accelerometers, hydrophones (underwater microphones), and acoustic emission instruments. Although its electrical resistivity and *RC* time constant remain quite high up to a T_c of 360°C (Table 4 and Fig. 8), its use is kept well under 200°C because of its tendency to age very rapidly, leading to depoling.[11]

5.3 Lead titanate

Lead titanate ($PbTiO_3$)(PT) the solid solution end member of PZT family, has a T_c of about 490°C. As in the case of PZT, a large number of modifications have been developed to optimize specific electrical and mechanical characteristics. Many commercial compositions of PT are doped with samarium or calcium for use in hydrophones, but this has the effect of lowering the T_c to around 240°C. Compositions, doped with other elements, have T_c values near 490°C and have found applications in knock sensors for automobile engines. The higher operating temperature of PT allows it to be mounted closer to the combustion chamber, thus giving a faster response time as compared to PZT.[6] The data presented in Tables 3 and 4 and Fig. 8 reflects the latter composition. Evaluation at 400°C has shown it to have a resistivity of only about 10^5 Ω-cm. Such a low value would adversely affect the *RC* time constant.

5.4 Lead metaniobate

Lead metaniobate ($PbNb_2O_6$)(PN) belongs to the tungsten-bronze family. Because of its low Q_m (wide bandwidth) and relatively high d_{33} to d_{31} ratio (high degree of anisotropy), PN finds its greatest use in transducers in NDT and medical diagnostic imaging. Commercial PN compositions are modified to enhance specific electrical characteristics but at the expense of the T_c. A commonly used composition contains about 10% Ba(BPN) and has a T_c of about 400°C. Although BPN is reported to resist depoling up to its T_c, limitations are imposed by its high conductivity above 300°C.[35] Figure 8 reveals that BPN exhibits the lowest resistivity of the materials tested. Other problems associated with this material are its high level of porosity and relatively low mechanical strength.[21]

5.5 Bismuth titanate

Bismuth titanate ($Bi_4Ti_3O_{12}$) is the titular compound of bismuth layer structure ferroelectrics (BLSF). Modification by one or more of a large number of other elements, to enhance dielectric and piezoelectric properties, is also common. A member of the family, reported to have favorable piezoelectric properties, high resistivity and high T_c (>600°C), is $Na_{0.5}Bi_{4.5}Ti_4O_{15}$, (NBT). Commercially available,[25] it is used in accelerometers operated at temperatures up to 400°C.[36] The strongest mode of vibration in NBT is the [33] mode but d_{33} and g_{33} are somewhat lower than many of the perovskite ferroelectrics previously discussed (see Table 3). Nevertheless, the high T_c and high resistivity make NBT an attractive, moderately high-temperature piezoelectric.

Although not commercially available, other complex variations of BLSF compounds have been reported to have Curie temperatures of over 800°C. Representative of this group is Bi_3TiNbO_9 for which room temperature values of d_{33}, and g_{33} were reported to be near those of NBT.[37] To date, little else has been reported about other properties, particularly at high temperature.

Unlike previously mentioned ferroelectric ceramics, BLSF materials can be made with grains having a plate-like structure. In ceramics formed and fired by conventional processes, these grains are oriented in a more or less random fashion, which leaves only a limited number of crystallographic orientation directions available for polarization due to the low symmetry of the structure. The achievement of an optimum degree of remnant polarization in polycrystalline ceramics necessitates the use of some mechanism to provide grain orientation. By employing hot forging techniques, researchers have prepared samples of several BLSF family members which display a high degree of texturing. The textured samples exhibited a twofold increase in coupling coefficient k_{33} and piezoelectric constant d_{33} over those from conventional sintering.[37]

5.6 Lithium niobate

Lithium niobate ($LiNbO_3$)(LN) has the corundum structure and a reported Curie point near 1150°C. Single crystals are grown from a melt using the Czochralski technique. Single crystals are preferred because of the higher piezoactivity, as well as difficulties encountered in conventional sintering of the polycrystalline form. As with polycrystalline ferroelectrics, single crystals of $LiNbO_3$ exhibit a multidomain structure, and must be polarized. This is accomplished by applying relatively small electric field (1 V/cm DC) at a temperature just below the T_c, thus converting the structure to single domain. The polarized crystal is then sliced along the desired axis indicated for the application, and the faces polished.[38] For accelerometers, electrodes are usually applied parallel to the poling axis to take advantage of the greater value of the d_{15} piezoelectric constant and to eliminate pyroelectric effects The voltage output of LN ($g_{15} = 91 \times 10^{-3}$ Vm/N) is significantly larger than those of the other piezoelectric materials listed in Table 3 due to the inherently low dielectric constant. Sensitivity remains high up to 400°C (Table 4) but resistivity is the limiting factor for use above 650°C (Fig. 8).

The tantalum analog, $LiTaO_3$, exhibits many of the same characteristics of $LiNbO_3$; however, the T_c (720°C) and piezoelectric constants are somewhat lower, thus offers no apparent advantage for high-temperature acoustic sensors.

6 NEW HIGH-TEMPERATURE PIEZOELECTRIC MATERIALS

In general, the ferroelectric ceramics discussed thus far are limited to temperatures of approximately $T_c/2$. Therefore, applications requiring yet higher temperatures than those just presented, require other materials such as ferroelectrics with higher Curie temperatures, oriented polar non-ferroelectric materials, non-polar piezoelectric single crystals, or piezoelectric thin films. Several novel materials currently under investigation are presented below.

6.1 Perovskite layer structure (PLS) ferroelectrics

PLS ferroelectrics have the general formula $A_2B_2O_7$ and possess an anisotropic layered structure similar to the BLSF family. Single crystals of the two most recognized compounds, $Sr_2Nb_2O_7$ and $La_2Ti_2O_7$, possess the highest known ferroelectric Curie temperatures, 1342°C and 1500°C respectively.[39,40] Thus, PLS ferroelectrics have been proposed for use in transducers with high operating temperatures and good thermal stability. Unfortunately, the cost of growing good-quality single crystals is high due to the high melting points of these compounds (which is considerably higher than that of $LiNbO_3$).

In a manner similar to that used for BLSF compounds, previously described, hot-forging was used to synthesize PLS compounds. The resulting samples, shown in Fig. 9, were found to exhibit near theoretical density and high degree of orientation, and polarizability.[29,41] The use of solid solutions with $Sr_2Ta_2O_7$(T_c = −107°C), to adjust T_c of the parent compound to <900°C, aided poling and the subsequent piezoactivity. One compound, $Sr_2(Nb_{0\cdot5}Ta_{0\cdot5})_2O_7$, had a T_c of 820°C and demonstrated the ability to resist depoling at temperatures as high as 650°C. The electromechanical properties for $Sr_2(Nb_{0\cdot5}Ta_{0\cdot5})_2O_7$ are listed in Table 4. Although the room temperature *d*-coefficient is low ($d_{24} = 2{\cdot}6 \times 10^{-12}$ C/N), a value similar to that of quartz, the piezoelectric voltage constant at 400°C is significant ($g_{24} = 6{\cdot}3 \times 10^{-3}$ Vm/N). From Fig. 8 is is also seen that the hot-forged PLS ceramics have the highest resistivity at elevated temperatures of any of the materials tested. By using compositions of $Sr_2(Nb_{1-x}Ta_x)_2O_7$ with a higher Nb content, the T_c can be increased to 1200°C with no loss of resistivity. Efforts to optimize the composition and processing variables are currently under way and may lead to improved properties.[42]

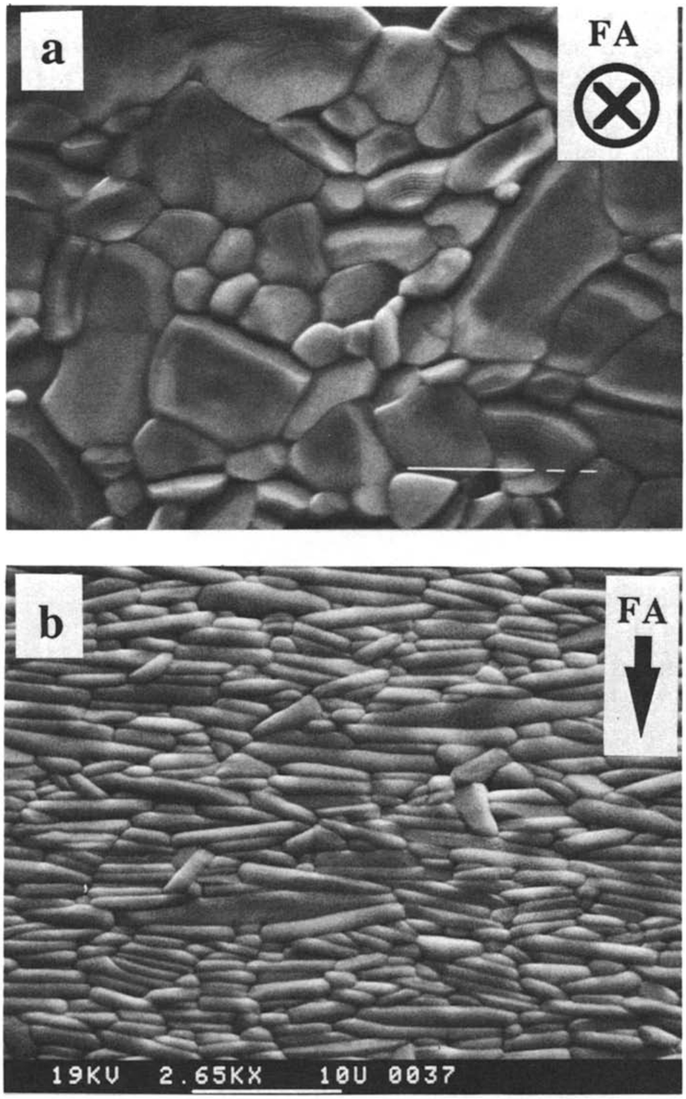

Fig. 9. Micrographs of hot forged $La_2Ti_2O_7$ ceramic. The plane perpendicular to the forging axis (FA) is shown in (a). The plane parallel to FA is shown in (b).

6.2 Polar glass ceramics

A classification of piezoelectric materials, totally diffrent from any previously discussed, exists in the form of polar glass ceramics. These non-ferroelectric compounds require single crystal growth or polar texturing for piezoactivity. One successful technique for obtaining oriented grains is the texturing of microscopic recrystallized polar crystals in an amorphous glassy matrix. Using a controlled thermal gradient technique, the polar material $Ba_2TiSi_2O_8$ (fresonite) was recrystallized from a glass of the composition $2BaO3SiO_2TiO_2$ resulting microstructure that contained polar oriented grains. With room temperature values of d_{33} and g_{33} were 7×10^{-12} C/N and 88×10^{-3} Vm/N respectively, and relatively low density (4·01 g/cm^3), these composite materials may be of interest for hydrophone applications.[43]

It has been suggested that polar glass ceramics may also be useful for high temperature piezoelectric devices. Since they are polar, but not ferroelectric, poling is unnecessary and, consequently, there is no problem with depoling or aging effects. Foreseen problems do exist, though, including softening of the glassy phase and possible ferroelastic phase transitions that would restrict the high-temperature capabilities. Further evaluation of the electrical properties, including resistivity at high temperatures, is needed before these materials can be commercialized.

6.3 AlN thin films

In recent years, numerous studies have been made of the piezoelectric properties of thin films of several materials, including the ferroelectrics previously discussed.[21] Thin films of non-ferroelectric materials are also of great interest, including aluminium nitride (AlN). Because of its exceptionally high thermal conductivity and dielectric breakdown strength, polycrystalline (AlN) is an important ceramic material used in substrates for hybrid microelectronics, but in the bulk form exhibits no piezoelectric activity. However, when properly oriented on a compatible substrate, AlN thin films exhibit piezoelectric properties which have been studied for their potential use as pressure transducers, speakers, and SAW devices.[44]

Of most interest for this report are the high-temperature piezoelectric properties of AlN thin films. A recent paper[45] reported that an AlN SAW device deposited on a fused quartz substrate, operating at 60–100 MHz, exhibited piezoelectric responses at temperatures up to 1150°C. Other reports on chemical vapor deposited (MOCVD) AlN thin films have shown room temperature d_{33} values of $5{\cdot}5 \times 10^{-12}$ Vm/N (about the same as $LiNbO_3$) and a dielectric constant K_{33} of 12.[46] Tsubouchi and Mikosheba[47] reported room temperature resistivity values of 10^{16} Ω-cm, a value higher than any other piezoelectric material discussed in this paper. With this combination of very high temperature operation, high resistivity, and reasonable piezoelectric coefficients, it seems that AlN thin films warrant further investigation.

7 SUMMARY

High-temperature technology is of major importance for chemical and material processing, automotive, aerospace, and power generating industries to name just a few. For many, the primary benefit of operating at higher temperatures is the direct cost savings associated with increased efficiency in fuel conversion. In a related matter, the development of advanced structural materials such as silicon nitride and carbon–carbon composites promotes a need for high-temperature electronic materials to monitor processing of these systems. This is exemplified by the recent organization of the First International High Temperature Electronics Conference by Sandia National Laboratory and Wright Laboratory.[48] Along with semiconductor, capacitor, magnetic, and packaging materials, electromechanical transducing materials are required to sense strains, vibrations, and noise under severe thermal conditions. Of the several different types of acoustic and strain sensors investigated, including accelerometers, strain gauge, air gap, eddy current, buffer rod, and fiber optic, piezoelectric types offer the best candidates when one considers sensitivity, cost, and design.

Figure 10 summarizes the maximum use temperature of widely commercial piezoelectric materials and the projected values of two experimental ones. If an operating temperature of 400°C or greater is required, the number of available sensor materials is clearly limited. If an operating

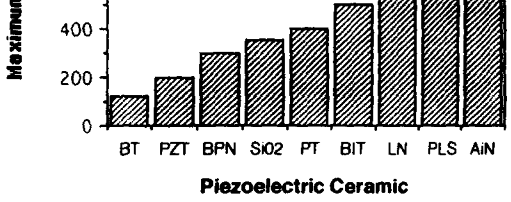

Fig. 10. Approximate maximum temperature of operation for piezoelectric ceramics. Most are estimated by combined consideration of the ferroelectric Curie temperature, sensitivity, and measured electrical resistivity. Others are known with a higher degree of certainty such as barium titanate (BT) limited by its T_c of 125°C, lead zirconate titanate (PZT) known to experience accelerated depoling at 200°C and quartz (SiO_2) with a maximum use temperature of 350°C.

temperature 750°C is chosen, there is no commercial material available. It is reasonable to assume that there now exists a need for vibration sensors that can function at 750°C, or even at 1000°C, and that the need will be even more pressing in the future. The design of such sensors presents a great challenge and will require the development of new materials and novel processing techniques.

This paper has discussed two options for very high temperature piezoelectric materials. The first was the family of PLS ferroelectrics, with the highest known T_c (some greater than 1500°C), The second was non-ferroelectric AlN thin film with a reported operating temperature of 1150°C. New ferroelectric compounds, also with very high T_c, have been predicted based on the Abrahams–Kurtz–Jameson relationship and an extensive inorganic crystal structure database.[49] This study, and other computer assisted studies of materials, may well identify the next generation of high-temperature vibration sensor materials.

ACKNOWLEDGMENT

The authors wish to thank NASA-Lewis for their support under grant number NA63-125.

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