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SHORT COMMUNICATION

Manufacturing Technology of Lead Zirconate Titanate Cylindrical Elements for Passive Transducer Arrays

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ABSTRACT

State-of-the-art technology has been developed for the fabrication of 33 mm hollow cylindrical elements from Lanthanum-doped lead zirconate titanate-based material suitable for passive surveillance arrays of SONAR systems. It covers properties of the material composition, isostatic pressing technique, precision machining, sintering to produce dielectrically sound distortion-free cylindrical elements, ceramic grinding, electroding, poling to achieve electromechanical properties, and evaluation of dielectric, piezoelectric, and elastic properties of the cylinders.

Keywords: PZT, lead zirconate titanate, piezoceramics, passive transducer arrays

NOMENCLATURE

- □ Element from lanthanide series
- d_{33} Piezoelectric charge constant
- d_{31} Piezoelectric charge constant measured at right angle to poling direction
- g_{31} Piezoelectric voltage constant
- L length of the element
- fr Resonance frequency
- fa Anti-resonance frequency
- Q_m Mechanical quality factor
- Z_m Impedence at fr
- C_o Static capacitance
- S_{11}^E Compliance at constant field
- K_3^T Dielectric constant
- K_{31} Coupling factor far below mechanical resonance, where electrodes are perpendicular to three axes and stress or strain is in one direction
- K_p Coupling factor, where electrodes are perpendicular to three axes and stress or strain is equal in all directions perpendicular to three axes

1. INTRODUCTION

Lanthanum-doped lead zirconate titanate piezoceramics are excellent energy converters. These enegy convertors are widely used for several strategic applications like impact initiation of hollow charge warheads and naval systems. These also find applications in industrial ultrasonics and medical diagnostic equipment.

Among the various lead zirconate titanate grades, lanthanum-doped lead zirconate titanate material have high dielectric constant, high piezoelectric charge coefficient, intermediate Curie point, and

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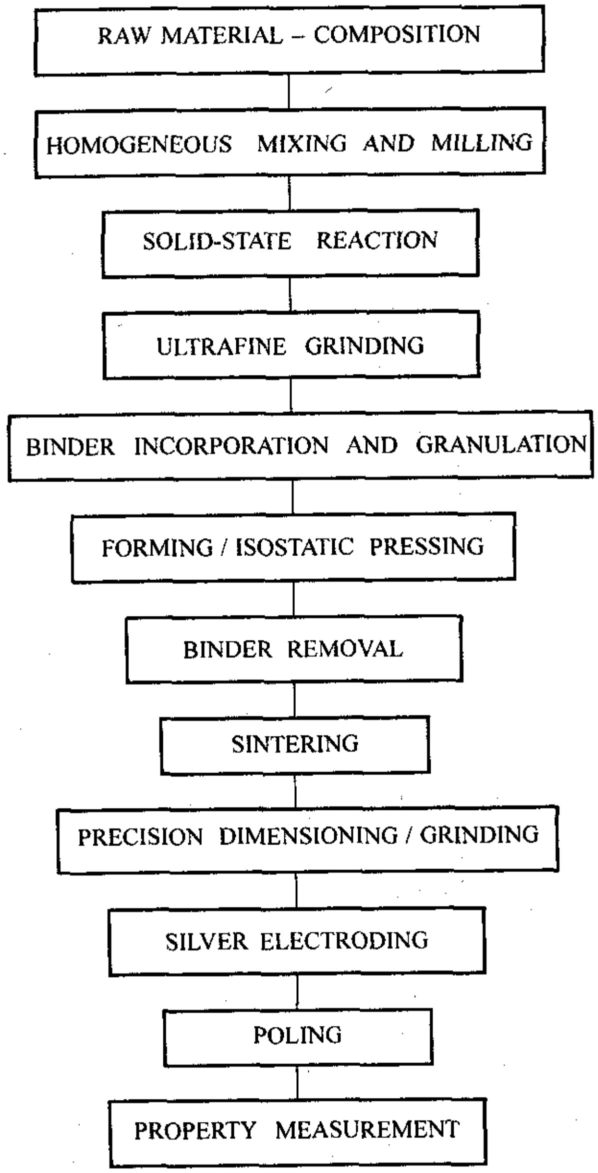

Figure 1. Process flow chart

high sensitivity, useful for receiving-type underwater transducer applications. Though the composition of this material is known[1], converting it into the required shape and size involves sophisticated technology, optimising several interdependent process parameters so as to obtain finished product with requisite electromechanical properties.

Here, the technology of manufacturing 33 mm diameter lanthanum-doped lead zirconate titanate hollow cylinders at pilot plant scale has been explained, including the technique established and the results obtained.

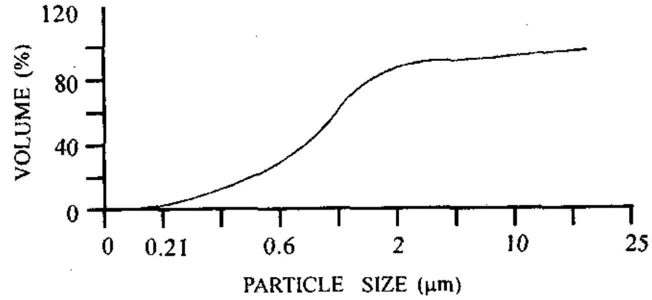

Figure 2. Typical particle size distribution

2. PREPARATION OF POWDER COMPOSITION

Lead zirconate titanate powder was prepared through mixed oxide route as per schematic flow chart (Fig. 1). Five kilogram batches having tentative formula $Pb_x \square_{1-x} (Zr_y\ Ti_{1-y})\ O_3$,[1,2] where x varies from 2 to 7 and y is adjusted to morphotropic phase boundary, were made out of oxides. Lead can be balanced with any element from lanthanide series and zirconium: Titanium ratio can be balanced with lanthanum (La). Properties corresponding to this composition are shown in Table 1. Solid-state reaction between the constituent elements PbO, ZrO_2, TiO_2 and La_2O_3 was carried out among 450 °C and 1000 °C. The calcined cakes were pulverised in vibro-energy mill to an average particle size of 1.3 μm. The particle size distribution[3] was checked

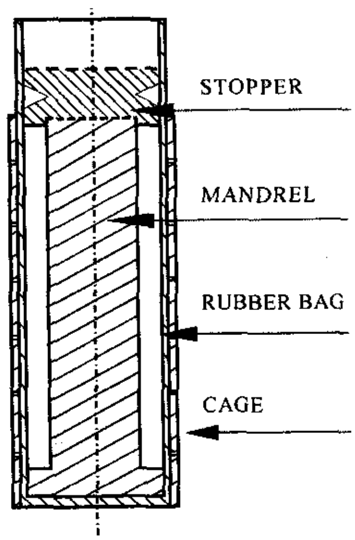

Figure 3. Wet bag isostatic pressing

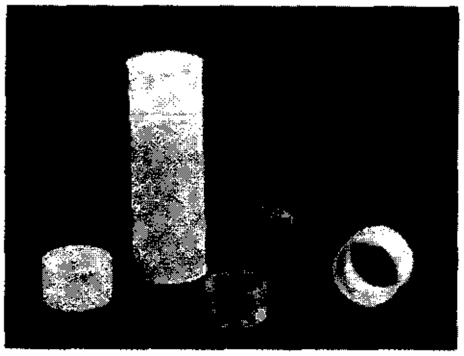

Figure 4. Isostatically pressed and sintered cylinders

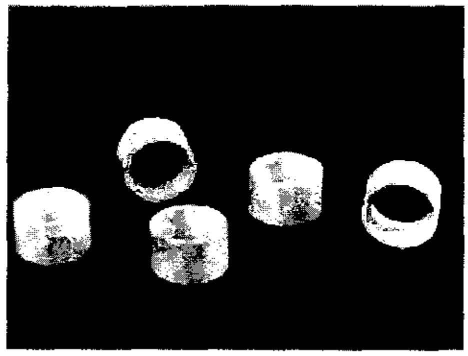

Figure 6. Finished product

by Malvern Mastersizer 2000MU equipment. A typical curve obtained is shown in Fig. 2. The batch properties in respect of dielectric and piezoelectric parameters were measured on 20 mm diameter × 1.5 mm thick discs[1].

3. FABRICATION OF CYLINDRICAL ELEMENTS

Cylinders were pressed using wet bag isostatic technique[2] (Fig. 3). Steel mandrel designed to give approximate sintered dimensions, considering the shrinkage of the powder, was placed inside the rubber bag. The assembly of the mandrel and the rubber bag was held by a steel cage. Granulated powder was filled in the rubber bag. The mould assembly was placed inside the pressure vessel of cold isostatic press and pressurised to 172.5 MPa. The powder compacts were extracted and heated to 600 °C to gain strength. Latter, these were machined on lathe machine for outer contour. Figure 4 shows pressed and machined cylinders subjected to time-temperature controlled sintering process.

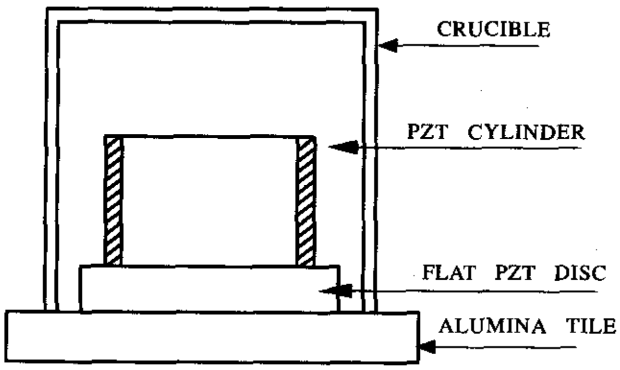

Figure 5. Sintering of PZT cylinder

The cylinders were sintered at 1240 °C for four hours in covered alumina crucibles under controlled lead oxide atmosphere[4]. The rate of heating was maintained between 4 °C/min to 5 °C/min with the help of programmable temperature controller. The sintering was optimised by adjusting thermal conditions, which produced uniform density, and high dielectric strength with improved piezoelectric properties. Fig. 5 shows the method employed for sintering of cylinders.

The technique used for sintering produced minimal distortion and was adopted for small-scale production. Ceramic grinding was performed on sintered cylinders to get required dimensions. Components were electroded with fired on silver paste. These components were then immersed in silicon oil bath and polarised by applying 2 kV/mm field. Figure 6 shows the finished product.

4. MEASUREMENT OF PIEZOELECTRIC PROPERTIES

Piezoelectric properties were measured after ten days of poling. Capacitance and tan δ were measured at 1 kHz using HP6462A LCR bridge. Dielectric constant (K_3^T) was computed using following standard formula:

$$K_3^T = C \times \text{Thickness} / (\text{Area permitivity of free space})$$

Resonance frequency, anti-resonance frequency, and impedance at resonance (Z_m), were measured

Table 1. Properties of five batches

Properties	Low signal data				
	Batch 1	Batch 2	Batch 3	Batch 4	Batch 5
Dielectric constant	2340	2320	2314	2357	2349
Tan δ	0.02	0.02	0.02	0.02	0.02
Piezo charge constant d_{33} (pC/N)	480	473	469	505	502
K_p	0.62	0.61	0.60	0.62	0.62
Density (g/cc)	7.48	7.45	7.46	7.48	7.48
Q_m	72	70	65	68	70

using HP4800A vector impedance meter. Piezoelectric charge constant (d_{33}) was measured using CPDT3330 Berlincourt d_{33} meter. Density (ρ) was measured using electronic weighing machine model CB-120 of Contech Instrument, having density determination kit. K_{31} and K_p were obtained from standard graphs[5,6]. Q_m, S_{11}^E, d_{31} and g_{31} were obtained from the following standard equations:

$$Q_m = fa^2 / [2\pi\ fr\ Z_m\ C_o\ (fa^2 - fr^2)]$$

where C_o is the static capacitance of the component measured at 1 kHz.

$$S_{11}^E = 1/\rho V_2^2$$

where V_2 is the sonic velocity ($2\pi\ fr\ l$).

$$d_{31} = K_{31}\sqrt{[\varepsilon_0\ K_3^T\ S_{11}^E]}$$

$$g_{31} = d_{31} / [\varepsilon_0\ K_3^T]$$

5. RESULTS & DISCUSSION

The average values of dielectric and piezoelectric properties measured on 10 numbers of 20 mm diameter × 1.5 mm thick discs for five batches, corresponding to composition $PbNb(ZrTi)O_3$ each of 5 kg, are given in Table 1. It has been observed that the variations in piezoelectric, dielectric, and elastic parameters were less than 5 per cent of the mean value.

Based on the achievements of desired electromechanical properties, these batches were further taken for manufacturing hollow cylinders of 33 mm diameter. The 1346 cylinders were evaluated for their properties. Their results are shown in Table. 2.

Table 2. Statistical data on 1346 cylinders

Properties	Mean value	± 2 %	± 5 %	± 7.5 %	± 10 %
Dielectric constant K_3^T	2316.16	57 %	94 %	99 %	99 %
Density(g/cc)	7.589	87 %	99 %	99 %	100 %
Poisons ratio	0.35	36 %	69 %	88 %	92 %
S_{11}^E 10^{-12} m²/N	15.2	68 %	98 %	99 %	99 %
K_{31}	0.308	36 %	78 %	93 %	96 %
$g_{31} \times 10^{-3}$ Vm/N	– 8.41	41 %	75 %	87 %	95 %

6. CONCLUSION

Hollow cylinders of 33 mm diameter were fabricated out of Lanthanum-doped lead zirconate titanate ceramic composition near morphotropic phase boundary, by mixed oxide route. Having precise control on the batch composition and process parameters, it was possible to achieve requisite electromechanical properties with rejections less than 5 per cent, maintaining the quality control in the production.

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Contributors

Mr PS Gaware joined DRDO at the Armament Research & Development Establishment (ARDE), Pune, in 1981. He has received the *DRDO Cash Award* for the development of indigeneous technology of the Navy-type V material and piezoceramic cylinders for SONAR system.

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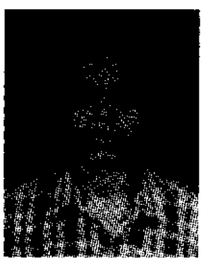

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