Def Sci J, Vol 41, No 4, October 1991, pp 399-408

Production Technology of Lead-Zirconate-Titanate Type-4 Spherical Elements for Underwater Transducers

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ABSTRACT

The paper describes the production technology evolved for the fabrication of 60 mm hollow spherical elements from lead zirconate titanate type-4 material, suitable for use in the manufacture of underwater omnidirectional transducers. It covers the characteristics of the starting powder, techniques of isostatic pressing, precision spherical machining, sintering to produce dielectrically sound, distortion-free hemispheres to the required physical dimensions, electroding, poling to achieve the inherent electromechanical properties, adhesive bonding of hemispheres and evaluation of ultimate dielectric and piezoelectric properties of the spheres.

1. INTRODUCTION

Most of the underwater transducers are electroacoustic in nature since other forms of energy except sound are heavily attenuated within short range and hence not much useful for many undersea applications. Underwater acoustic transducers are required for several Naval applications such as sonobuoys, torpedoes, SONARS, surveillance systems, towed arrays, etc.

The present day SONARS and hydrophones use polycrystalline ferroelectric ceramics, generally lead-zirconate-titanate (PZT) based materials. Amongst the various PZT grades[1], the moderately hard ferroelectric grade PZT type-4 is considered to be well suited for most of the applications because of its superior high signal parameters. It has much higher power handling capacity per unit volume than the conventional

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Received 21 May 1990, revised 26 December 1990

barium titanate material. Since this material has high Curie temperature, reasonably high electromechanical properties coupled with low electrical and mechanical losses, it can be used both as a sensor as well as radiating element in transducer designs. The technology of PZT type-4 material is already available locally, but for specified applications the shape and the size of the required elements are needed to be perfected by way of optimising several interdependent processing parameters so as to obtain the end product with highest electromechanical characteristics within the acceptable limits. In this context, establishing the technology of 60 mm dia PZT type-4 hollow spheres at pilot plant scale for certain specific transducer applications becomes relevant. This paper describes the techniques established and the results obtained.

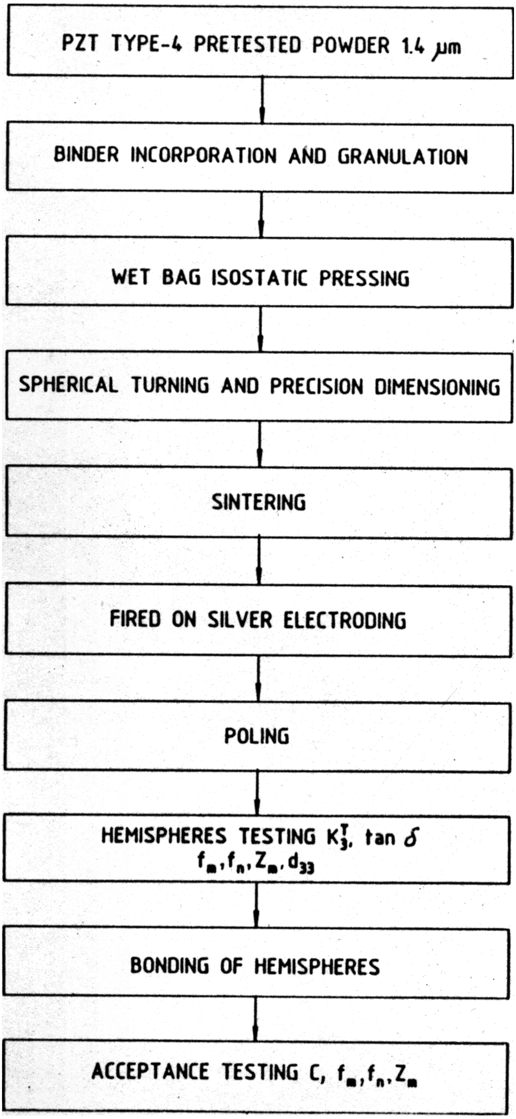

Figure 1 Flow chart for the production of hollow spheres.

Since the technology of PZT type-4 material with the electromechanical properties as shown in Table 1 has already been established[2], it was considered to work out the batch production processes of fabricating 60 mm OD and 3.2 mm wall thickness hemispheres from the proven lots of PZT powder by cold isostatic pressing and machining to precision physical dimensions, etc as per the schematic flow chart depicted in Fig. 1. The spheres are then assembled by adhesive bonding of the two poled hemispheres displaying the acceptable dielectric and piezoelectric properties, after interconnecting the internal electrodes and the electrical feed-through. The spheres, then are tested for physical and electromechanical properties against the laid down acceptance criterion.

2. EXPERIMENTAL WORK

2.1 PZT Powder Production

Batches consisting of 8 kg PZT material having the nominal formula $Pb_{0.94}\,Sr_{0.06}\,(Zr_{0.53},\,Ti_{0.47})\,O_3$ were made from the respective oxides/carbonates by the usual wet ball-milling and solid state reaction techniques. The calcined mass was pulverised in a fluid jet mill to an average particle size of 1.2 to 1.4 μm. The particle size distribution was checked by 5000 D Micromeritic Sedigraph equipment. A typical curve obtained is shown in Fig. 2. The surface area of this powder as measured by BET nitrogen adsorption was around 5.4 m^2/g. The quality of each batch in respect of relevant dielectric and piezoelectric parameters was checked by conducting measurements on 20 mm dia and 1.5 mm disk samples produced in the usual manner. In order to avoid spread in the properties of the finished elements, two such pre-tested batches were blended together to provide 16 kg of powder for use in the manufacture of hemispherical elements.

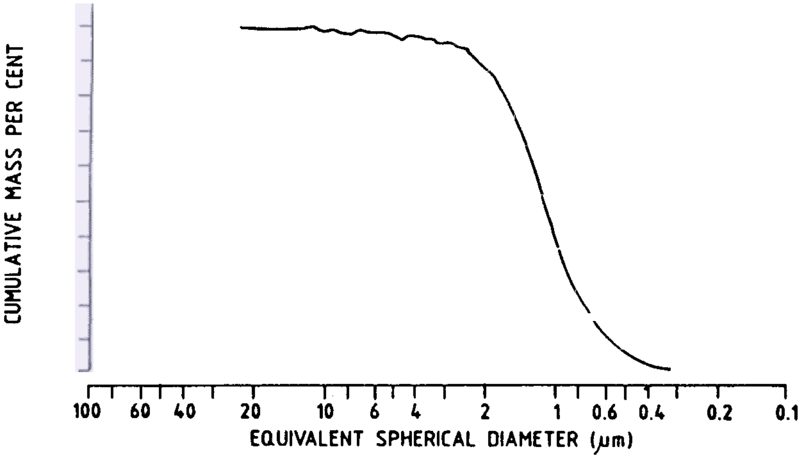

Figure 2. Typical particle size distribution.

2.2 Hemisphere Fabrication

The method of fabrication for green hemispheres had been accomplished with cold isostatic pressing of the PZT powder by using wet bag tooling as shown in Fig. 3. The internal spherical contour of the hemisphere to the net shape after allowing for the shrinkage in sintering had been achieved by using the steel core. The moulds during powder-filling were vibrated on shaker table, evacuated and then compressed at about 175 MPa pressure. The pressed pieces were dried and subsequently heated at 600 °C for 2 hr to consolidate the compressed bodies to gain strength. Later these were machined for the outer contour on a lathe machine with a spherical turning-tool attachment. The cutouts meant for the insertion of feed-through for electrical connections were manually done on each piece. Figure 4 shows the as-pressed and machined hemispheres which were then subjected to time- temperature-controlled binder burn-out treatment.

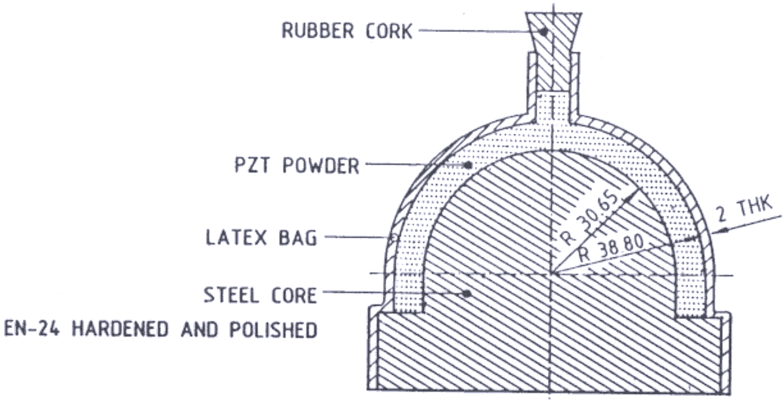

Figure 3. Wet bag tooling for cold isostatic pressing of 60 mm dia hemispheres.

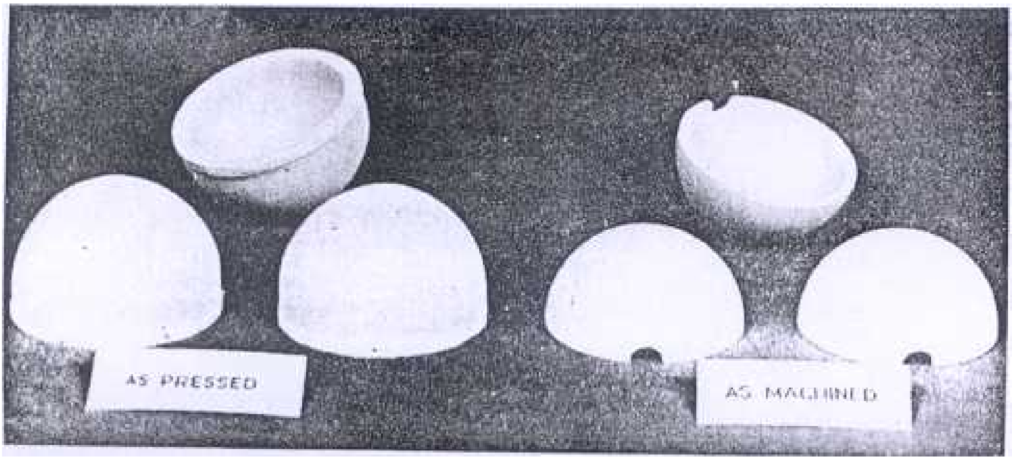

Figure 4. As-pressed and machined hemispheres.

2.3 Sintering, Electroding and Poling

The hemispheres were sintered at 1260 °C for 3 hr in covered alumina saggers under controlled PbO atmosphere emanating from PbO-enriched lead zirconate pellets placed therein. Controlled rate of heating and cooling were adopted with the help of programmable temperature controller which assured the required precision in the thermal treatment and reproducibility of the sintering process. The optimisation of the sintering process revolved around the adjustment of thermal conditions which produced the highest density along with high dielectric strength and low dc conductivity to facilitate poling at the elevated temperatures with least dielectric failure and improved piezoelectric properties. Scanning electron micrograph of the representative sintered sample is shown in Fig. 5.

One of the techniques to be established in the sintering process was to achieve uniform shrinkage of the hemispheres while maintaining its sphericity. Several sintering runs were made with different orientations of the green hemispheres on the supporting rings/disks (Fig. 6), but finally the inverted setting of the hemisphere on a flat disk (Fig. 6(c)) produced minimal distortion and has been adopted for the regular production. After firing, the pieces were lightly polished with fine emery abrasive and the final check on the dimensions was carried out. Then, the pieces were silver electroded with fired-on silver preparation which had to be especially evolved for the purpose by critically adjusting its glass content and curing cycle to provide the requisite bond strength of the electrode-PZT substrate. These elements while immersed in silicon oil, were then poled at about 100 °C for 30 min with 10-11 kV electric potential applied across the thickness.

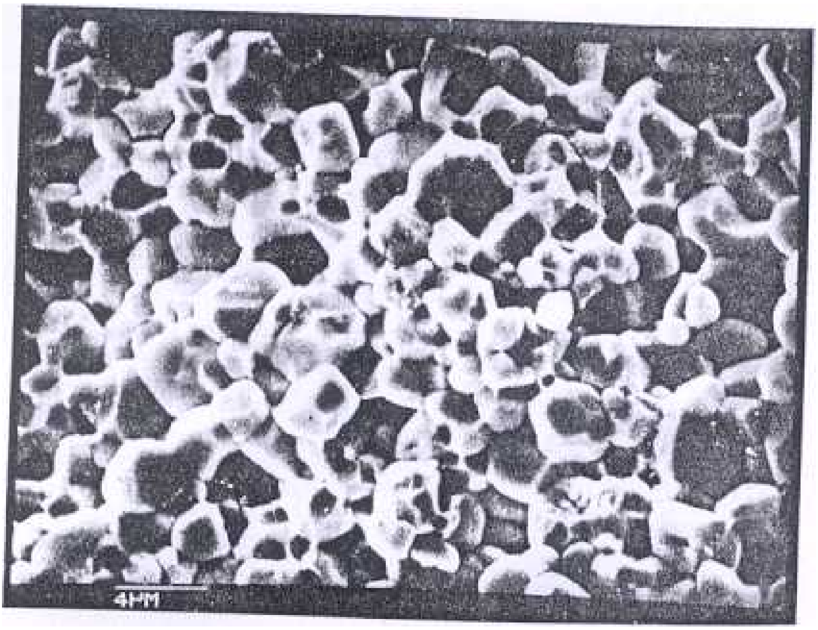

Figure 5. Scanning electron micrograph of the sintered PZT type-4.

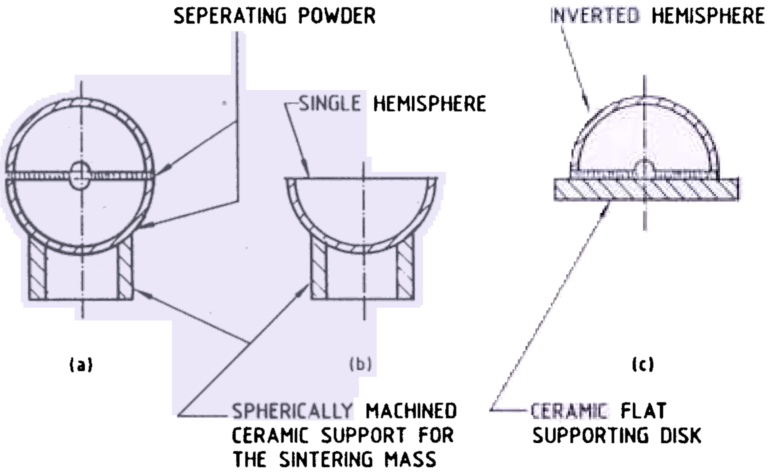

Figure 6. Method of sintering hemispheres.

2.4 Dielectric and Piezoelectric Properties

After ten days of poling, the hemispheres were checked for their dielectric constant and tan δ at 1 kHz low field with the help of LCR meter HP 6462A. Resonance and anti-resonance frequencies and impedance at resonance were checked with HP 4800A Vector Impedance Meter.

2.5 Fabrication and Evaluation of Spheres

Hemispheres with closely matching electronic properties and physical dimensions were selectively chosen from the given lot for adhesive bonding to produce structurally sound spherical elements. Necessary lead wires were soldered to the two inner electrodes which in turn were connected to the feed-through. The feed-through and the hemispheres were bonded with high temperature Ciba epoxy resin. Figures 7 and 8

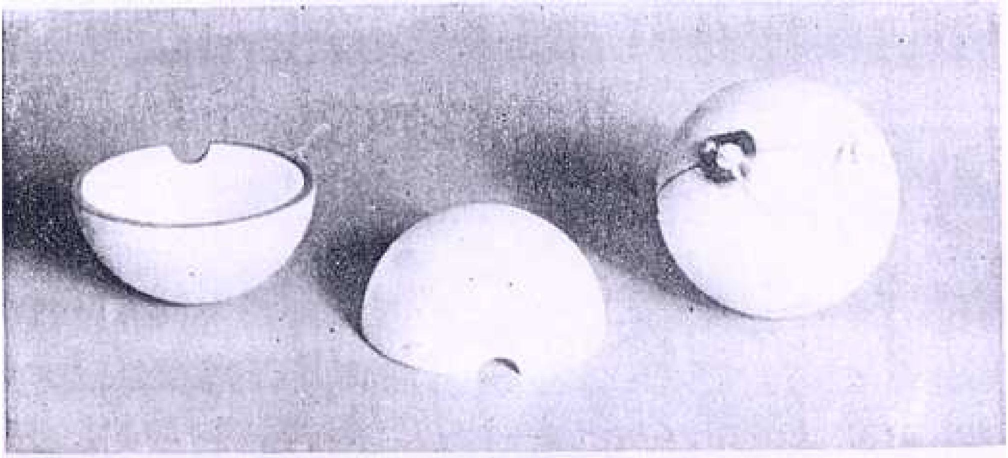

Figure 7. Assembled spheres with hemispherical elements.

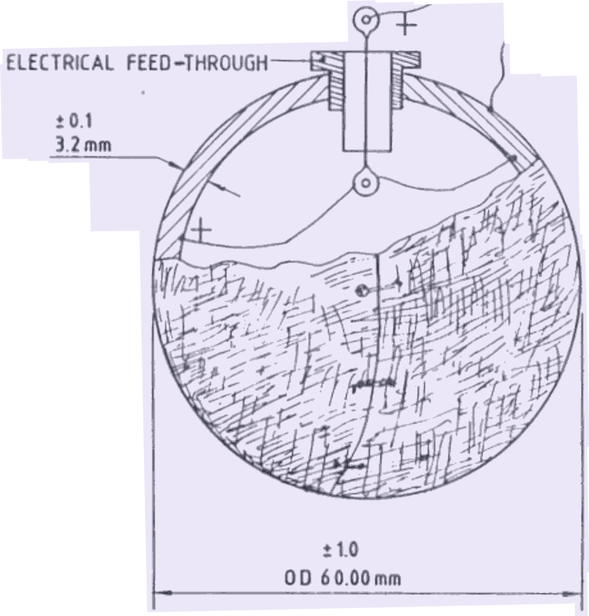

Figure 8. **Engineering sketch of the assembled sphere.**

depict the assembled spheres. The assembled spheres were then checked for acceptance testing by way of measurement of capacitance (C), electrical dissipation factor (tan δ), frequency of maximum admittance (f_m), frequency of minimum admittance (f_n) and impedance at the frequency of maximum admittance (Z_m).

3. RESULTS AND DISCUSSION

3.1 Reproducibility of PZT Powder Batches

In the effort to achieve uniform quality and reliability in the finished spheres, it became a prerequisite to start with PZT powders originating from different lots that should have the inherent quality to impart excellent dielectric and piezoelectric properties. This check has been exercised, as a matter of routine, on 16 kg lots of the powder by testing atleast 10 disks duly produced as explained in Section 2.1, before the given lot is released for production. Desirable physical and electromechanical properties of PZT type-4 are shown in Table 1. Pertinent data in respect of dielectric constant (K_3^T), dissipation factor (tan δ), charge coefficient (d_{33}), radial coupling constant (k_p), and mechanical quality factor (Q_m) from 10 such randomly selected powder batches is shown in Table 2 the values of which closely match with the desired values shown in Table 1. The variation are little and within close tolerances. This type of monitoring assured that a close control, critical analysis and modifications could be exercised in the desired direction on several interdependent processing variables to attain reasonably good electromechanical properties in the end product with good process efficiency.

Table 1. Desirable physical and electromechanical properties of PZT type-4 material

Sintered density	7.60 g/cc
Curie temperature	320 °C
Relative dielectric constant (K_3^T) at 1 kHz low field	1100–1400
Dissipation factor (tan δ) at 1 kHz, low field	0.005 (max)
Piezoelectric voltage coefficient (g_{33})	25×10^{-3} Vm/N Nom
Piezoelectric charge coefficient (d_{33})	285×10^{-12} C/N Nom
Radial coupling constant (k_p)	0.52–0.60
Mechanical quality factor (Q_m)	500 (min)

Table 2. Reproducibility of PZT type-4 batches

Batch No.	K_3^T	tan δ	k_p	d_{33} ($\times 10^{-12}$ C/N)	Q_m
6	1373	0.004	0.55	300	530
7	1364	0.004	0.56	295	540
9	1370	0.004	0.57	298	536
11	1365	0.003	0.57	295	542
14	1360	0.004	0.56	290	560
16	1350	0.003	0.56	296	545
18	1368	0.003	0.55	297	560
20	1363	0.003	0.55	285	558
22	1370	0.003	0.56	297	540
24	1360	0.003	0.57	293	561

3.2 Physical and Electromechanical Properties of the Assembled Spheres

Physical data on the poled and assembled spheres, taken from 10 different batches is given in Table 3. It may be inferred that the material of the spheres had been sintered to a density $\geqslant 7.6$ g/cc with good consistency and to a fine grain structure as depicted in Fig. 5. The material acquired reasonably good dielectric strength to facilitate poling of comparatively a large surface area of sphere bodies at elevated temperatures with the electric field of the order of 32 kV/cm to realise the inherent piezoelectric properties of this material with good yield. Also, the distortion on the diameter has been controlled within ± 0.25 mm, without resorting to any expensive post-sintering material removal techniques. Low signal relative dielectric constant (K_3^T) and electrical loss factor (tan δ) are in general agreement with this class of materials[3]. A fair uniformity is reflected in the frequencies at minimum impedance (f_m) and maximum impedance (f_n) and the corresponding value of k_{eff}. The figures of the impedance of $f_m(Z_m)$ also present fair uniformity between 1.1 to 1.7 ohms which is the result of careful practices adopted in the selective assemblies and bonding of the hemispheres. In the preliminary examination for the transducer devices, these spheres have been found to function satisfactorily. However, there is still some scope to improve the k_{eff} and studies are in hand to evaluate the impact of several process variables, especially the effect of the characteristics of starting PZT powder, sintering, poling techniques, etc on this parameter.

Table 3. Physical and electromechanical properties of assembled spheres

Batch No.	Sintered density (g/cc)	Average OD (mm)	Maximum observed distortion on OD (mm)	K_3^T	tan δ	Radial mode frequency (kHz) f_m	f_n	Z_m (ohm)	k_{eff}
17	7.64	60.04	0.46	1339	0.002	31.51	36.25	1.20	0.49
24	7.68	60.40	0.44	1343	0.002	31.53	36.24	1.10	0.49
45	7.66	59.90	0.40	1304	0.002	31.50	36.16	1.55	0.49
52	7.62	60.56	0.34	1307	0.002	31.50	36.55	1.10	0.51
54	7.70	59.92	0.50	1308	0.002	31.51	36.42	1.20	0.50
57	7.68	59.82	0.34	1315	0.002	31.25	36.60	1.20	0.52
59	7.67	60.11	0.42	1299	0.002	31.30	36.70	1.20	0.52
	7.63	59.95	0.32	1262	0.002	31.35	36.64	1.30	0.52
73	7.60	60.00	0.47	1279	0.002	31.10	36.52	1.68	0.52
77	7.65	60.16	0.28	1277	0.002	31.20	36.58	1.40	0.52

Table 4. Statistical analysis of the properties of spheres

Statistical parameter	Capacitance (nF)	f_m (kHz)	k_{eff}
Number of samples	50	50	50
Minimum value	35.300	30.900	0.460
Maximum value	39.200	31.520	0.510
Mean $\bar{x}$	37.400	31.152	0.480
Mean deviation (MD)	0.700	0.132	0.008
Out of 3 MD limits	Nil	2	5
Standard deviation	0.865	0.162	0.009
Lower limit ($\bar{x}$ + 3 MD)	35.300	30.757	0.456
Upper limit ($\bar{x}$ + 3 MD)	39.500	31.547	0.504
% variation from the mean	± 5.60	± 1.27	± 5.00

3.3 Reproducibility of Production

Statistical analysis on a sample of 50 spheres randomly selected from the accepted production lots is given in Table 4. It may be seen that a tolerance of ± 5.6 per cent, ± 1.27 per cent and ± 5.0 per cent had been achieved in respect of capacitance, f_m and k_{eff} respectively while working on the batch production of these spheres on pilot plant scale. This had been possible with precise control on the PZT powder production and subsequent critical processing conditions with a view to produce spherical piezoelements which may function in transducer arrays wherein large variations in properties are not acceptable.

4. CONCLUSIONS

From the foregoing it is concluded that the pilot plant productionisation of hollow PZT type-4 hemispheres of 60 mm dia and 3.2 mm wall thickness with the requisite electrical connections, suitable for underwater transducer applications has been accomplished from 16 kg PZT type-4 powder batches of consistent quality and optimisation of all critical production parameters. All along, major emphasis has been on quality control aspects with a view to minimising variations in the physical dimensions and piezoelectric properties of the spheres with least rejections

ACKNOWLEDGEMENTS

The authors are thankful to Mr NS Venkatesan, Distinguished Scientist and Director, ARDE, Pune for the encouragement to pursue this work and for the permission to publish this paper.

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