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Piezoelectric Properties of Solid Solution Ceramics — A Note

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Abstract. Piezoelectric coupling coefficient for compositions in the quaternary system $(BaPb)(NbZr)O_3$ using Mason's technique are reported. It is found that the radial coupling coefficient is maximum for compositions at the morphotrophic boundaries in both tetragonal and orthorhombic phases of the system.

1. Introduction

The piezoelectric effect provides a connection between mechanical strain or stress and polarization or electric field. The crystals expand in the direction of the applied field and contract by a small amount in all directions perpendicular thereto. These characteristics are made use of for a variety of applications such as ultrasonic transducers and under-water sound communications. These require high mechanical Q and efficient electro-mechanical coupling.

It has been shown[1,2] earlier that compositions in the quaternary system $BaNb_2O_6 - PbNb_2O_6 - BaZrO_3 - PbZrO_3$ exhibit ferroelectric properties which get enhanced at the morphotrophic phase boundaries. This communication reports on the piezoelectric properties of the compositions crystallising with the tetragonal $PbTiO_3$ type and orthorhombic $PbNb_2O_6$ type phases.

2. Experimental

The raw materials ($BaCO_3$, PbO, Nb_2O_5, ZrO_2 all $\geqslant$ 98 % pure) were weighed in stoichiometric proportions and ground in an agate mortar under acetone. These were fired at 1280–1300°C for about two hours after a prefiring at 600–700°C for 12 hours, and the endproducts were pressed into disks of 12 mm diametre and 1-2 mm thickness in a Carver press at a pressure of 3,000 kg/cm^2. Fired on electrodes were applied.

The disks were polarized using a high tension transformer3 with a primary winding designed for 50 Hz, 200 (rms) a.c. and a secondary winding of 1500 volts (rms)

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a.c. The d.c. field across the disks works out to be of the order of 15 Kv/cm. The disks were dipped in silicone oil bath which was heated above Curie-temperature and the d.c. field applied after the temperature equilibrium was reached, and the sample cooled through the Curie-temperature. These were washed with petroleum ether and used for measurement of coupling coefficient based on the, Mason's method*. It is known that for simple modes of vibrations, the coupling factor K_r, resonance frequency and the anti-resonance frequency f_A are related. For example, in the case of a radially resonant disk polarized. along the axis, the relation can be expressed in the following form taking the electromechanical coupling coefficient as the ratio of the stored mechanical energy to total stored energy and the energy as proportional to the appropriate capacitor value in the simple equivalent circuit proposed for quartz by Van Dyke[5,6].

$$K_r^2 = 2\Delta f/f_R/\ 1 + 2\ \Delta\ f/f_R \qquad (1)$$

'where $\Delta f = f_A - f_R$

The disks were made part of a Colpitt Oscillator7 circuit and the resonance and the anti-resonance frequencies accurate to $\pm$ 0.5 kHz were determined by changing the tuning condenser. These were then used to calculate the radial coupling coefficient using the eqn. 1 above.

3. Results and Discussion

The values f_A, f_R and k_r alongwith the T_c (Curie-temperature) values for the compositions crystallising with the tetragonal $PbTiO_3$ and orthorhombic $PbNb_2O_6$ type phases is shown in Table 1 & 2. The highest values in both the phases are observed for compositions near the morphotrophic boundaries. The highest dielectric constant values (ϵ_{max}) are also observed for such composition?. Such type of behaviour is observed for compositions PZT-2 and PZT-4 also[8]. The present values of k_r compare well with the conventional commercial piezoelectric substances like LZ-4a (0.55, Brush Clevite Corporation, London)[9] and are higher than those reported for the $Pb(TiZr)\ 0$, system[10]. For example the composition $Pb(Ti_{0.47}Zr_{0.53})\ 0$, has a k_r value of 0.48.

Table 1. f_A, f_R and k_r values for compositions in the $PbTiO_3$ type phases

Composition	f_A	f_R	Δf	k_r	T_c (°C)
		kHz			
$Ba_{0.65}Pb_{0.075}Nb_{0.55}Zr_{0.45}O_3$	450.5	389	61.5	0.49	203
$Ba_{0.6}Pb_{0.1}Nb_{0.6}Zr_{0.4}O_3$	427.5	375	52.5	0.47	300
$Ba_{0.65}Pb_{0.05}Nb_{0.6}Zr_{0.4}O_3$	422	373.5	48.5	0.45	270
$Ba_{0.7}Pb_{0.0}Nb_{0.6}Zr_{0.4}O_3$	,419	375	44	0.43	248
$Ba_{0.6}Pb_{0.075}Nb_{0.65}Zr_{0.35}O_3$	456	389	67	0.51	209
$Ba_{0.55}Pb_{0.1}Nb_{0.7}Zr_{0.3}O_3$	461.5	394.5	67	0.50	224
$Ba_{0.6}Pb_{0.05}Nb_{0.7}Zr_{0.3}O_3$	422	370	52	0.47	220
$Ba_{0.65}Pb_{0.0}Nb_{0.7}Zr_{0.3}O_3$	417.5	367	50.5	0.46	210

Table 2. f_A, f_R and k_r values for compositions in the $PbNb_2O_6$ orthorhombic phase

Composition	f_A	f_R	Δf	k_r	T_c(°C)
	KHz				
$Ba_{0.1}Pb_{0.5}Nb_{0.8}Zr_{0.2}O_3$	466	396	70	0.51	214
$Ba_{0.1}Pb_{0.475}Nb_{0.85}Zr_{0.15}O_3$	511	422	89	0.54	229
$Ba_{0.05}Pb_{0.5}Nb_{0.9}Zr_{0.1}O_3$	519.5	411	108.5	0.59	252
$Ba_{0.1}Pb_{0.45}Nb_{0.9}Zr_{0.1}O_3$	456	394.5	61.5	0.49	247
$Ba_{0.15}Pb_{0.4}Nb_{0.9}Zr_{0.1}O_3$	400	356	44	0.44	224
$Ba_{0.2}Pb_{0.35}Nb_{0.8}Zr_{0.1}O_3$	496	416.5	79.5	0.52	204
$Ba_{0.1}Pb_{0.425}Nb_{0.95}Zr_{0.05}O_3$	461.5	394.5	67	0.50	198
$Ba_{0.2}Pb_{0.325}Nb_{0.95}Zr_{0.05}O_3$	456	394.5	61.5	0.49	180

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