# THE EFFECT OF GRAIN SIZE ON THE PROPERTIES OF PIEZOCERAMICS IN THE SYSTEM PbMg<sub>1/3</sub>Nb<sub>2/3</sub>O<sub>3</sub>-PbTiO<sub>3</sub>-PbZrO<sub>3</sub>

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Internal stresses arise during poling of piezoelectric transducers as a result of reorientation of the domain structure. If the grain size exceeds a certain critical limit, the internal stresses cause cracking at the grain boundaries. An expression for the critical grain size is derived in the present study. The formation of cracks by poling is determined in coarsegrained ceramics Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)<sub>0.44</sub>Ti<sub>0.44</sub>Zr<sub>0.12</sub>O<sub>3</sub>, doped with 1 wt. % MnO<sub>2</sub>. The presence of cracks is revealed by impaired mechanical strength and increased internal friction of the piezoelectric transducers.

#### INTRODUCTION

Piezoelectric transducers and resonators are made of substances exhibiting ferroelectric properties. Cooling of sintered ferroelectric ceramics from the high-temperature paraelectric phase to the low-temperature ferroelectric phase brings about formation, in the individual grains, of domains, that is regions showing mutual differences in the orientation of the spontaneous polarization vector.

The domain structure is formed in a way ensuring minimum internal stresses during the transition from the paraelectric phase to the ferroelectric one.

Changes in the domain structure occur, the domain walls being displaced, under the effect of the external DC electric field during poling. The domain structure is reoriented so as to align the directions of spontaneous polarization, allowed by the crystal symmetry, as closely as possible with the direction of the electric field introduced. Following discontinuation of the field, the domain structure does not regain reversibly its original arrangement, remaining partially reoriented. The poled specimen then shows piezoelectric properties.

The present study is concerned with the properties of piezoceramics in the ternary system  $Pb(Mg_{1/3}Nb_{2/3})O_3 - PbTiO_3 - PbZrO_3$ . The composition chosen was 44 mol %  $Pb(Mg_{1/3}Nb_{2/3})O_3$ , 44 mol %  $PbTiO_3$  and 12 mol %  $PbZrO_3$  doped with 1 wt. %  $MnO_2$ . Piezoceramic transducers of this composition exhibit superior values of the mechanical quality factor  $Q_M$  and high temperature stability of the resonance frequency,  $f_r$  [1].

In the phase diagram of the given system (Fig. 1), the composition of the solid solution in question is designated by the point F. This composition, close to the phase boundary between the tetragonal and the pseudocubic phases, has been chosen primarily with respect to attaining the required temperature stability  $f_r$  [1, 2]. In ceramics of the given composition, the cubic paraelectric phase is converted

to the tetragonal ferroelectric one at temperatures of about 220 °C. In the ferroelectric phase, the individual grains contain domains which are mutually separated by 180-degree and 90-degree walls.



Fig. 1. Phase diagram of the system  $Pb(Mg_{1/3}Nb_{2/3})O_3 - PbTiO_3 - PbT_iO_3$ , T - tetragonal phase, R - rhombohedral phase, PC - pseudocubic phase. The composition of the solid solution used in the present study is designated F.

Using small variations in the content of the initial components, it was possible to prepare the piezoceramic material in question either in coarse-grained or fine-grained forms. The properties of piezoceramic transducers of close compositions but with various grain sizes are compared in the present study. It was shown that if the grain size exceeds a certain critical value, cracks are formed at the grain boundaries as a result of internal stresses brought about by reorientation of the domain structure.

#### EXPERIMENTAL

The piezoceramic material of the given composition was prepared in the conventional way, i.e. by grinding the initial mixture of oxides, subsequent calcination and sintering. The differences in the grain sizes of the sintered transducers were achieved by varying the content of lead oxide in the raw material mixture. Two types of this ceramic material were prepared:

a) Fine-grained ceramics from a mixture of PbO, MgO, Nb<sub>2</sub>O<sub>5</sub>, TiO<sub>2</sub> and ZrO<sub>2</sub> corresponding to the stoichiometric composition  $Pb(Mg_{1/3}Nb_{2/3})_{0.44}Ti_{0.44}Zr_{0.12}O_3$  and with an addition of 1 wt. % MnO<sub>2</sub>.

b) Coarse-grained ceramics was prepared from a mixture of the oxides in which the PbO content was raised by  $3 \mod \%$  compared to the stoichiometric composition.

The weighed mixtures of oxides were wet-ground and homogenized, and after drying calcined at 900 °C. The calcined material was wet-ground, dried, granulated and press-moulded into platelets  $16 \times 6 \times 2.5$  mm in size.

The specimens of both compositions were sintered at 1220 °C for 1 hour, placed separately in platinum containers. The sintering was carried out in PbO atmosphere so as to suppress volatilization of this oxide from the mouldings.

The sintered specimens were ground to the dimensions of  $14 \times 4 \times 1$  mm, and over an area of  $14 \times 4$  mm metal-plated with silver-plating suspension. The transducers prepared in this way were then poled with a DC field of 0.8 to 3.2 kV/mm intensity for 30 minutes at 100 °C. In the course of poling the transducers were kept in silicone oil. Both the poled and unpoled specimens were tested for bending strength using a three-point fixture in the Instron 1122 tester.

According to the IRE standard [3], the frequency characteristics of the poled transducers were determined in the frequency region of the basic longitudinal oscillation. The mechanical quality factor  $Q_{\rm M}$  was determined with the transducers in the  $\pi$  circuit.

The structure of fracture surfaces of transducers after the bending strength tests was studied by means of the scanning electron microscope. Optical microscopy was used in evaluating the polished sections of both poled and unpoled transducers. The mean grain size in both types of the ceramic material was determined on micrographs of etched polished sections by Müller's method [4] which is based on finding the number of grains intersected by a system of parallel lines and establishing the mean length of secants  $d_s$ . The mean grain size was defined as  $d = 1.5 \times d_s$ . The mean grain sizes established for the fine- and coarse-grained ceramics were 6 and 16 µm respectively.

# Analysis of internal stresses in piezoelectric ceramics

Before discussing the experimental results, an evaluation will be made of the effect of internal stresses on the mechanical properties of the materials.

Internal stresses in ceramic materials can arise as a result of an anisotropy of thermal expansion during their cooling down from the sintering temperature. However, the piczoceramics in question is cubic above Curie temperature, its thermal expansion is isotropic, so that no internal thermal stresses can occur.

The cooling to Curie temperature brings about transition to the ferroelectric tetragonal phase. Permanent elongation amounting to  $e_{T_3} = (c/a) - 1 \approx 1.8 \times 10^{-2}$  takes place in the direction of spontaneous polarization i.e. in the direction of axis c of the crystal lattice. In view of the small change in the volume involved, permanent contraction  $e_{T_1} = e_{T_2} \approx -9 \times 10^{-3}$  occurs in the directions perpendicular to axis c. Such deformations would not bring about any internal stresses in a one-domain free grain. However, certain internal stresses arise in polycrystalline ceramics as a result of interactions with the neighbouring grains.

If the transition of the paraelectric phase into the ferroelectric one takes place on cooling down and without the effect of an external electric field, the considerable variability in the formation of the domain structure will play its part. The variability is due to the existence of three type  $\langle 001 \rangle$  directions and thus six possible directions of the spontaneous polarization vector in each grain. The formation of the domain structure will thus facilitate mutual adjustment of the grain shapes mostly through free deformation, without bringing about any major additional elastic deformation which would result in internal stresses. The piezoceramics in question did not exhibit any distinct impairing of mechanical properties due to transition from the paraelectric phase to the ferroelectric one. The internal stresses can therefore be considered negligible.

However, this does not apply to the poled piezoceramic material whose domain structure has reoriented under the effect of the electric field. The domain structure in its initial state, showing minimum internal stresses, becomes forcefully reoriented to align the direction of spontaneous polarization in the domains with the direction of type  $\langle 001 \rangle$  containing the smallest angle with the direction of the external electric field.

In the case of low-intensity electric fields, the displacement affects primarily the 180-degree domain walls. Their displacements are not associated with any creation of internal stresses, as they do not involve changes in the orientation of the tetragonal crystal lattice; the shapes of the grains are not affected by this reorientation of the domain structure.

In high-intensity electric fields, which also cause 90-degree domain walls to displace, the spatial orientation of the tetragonal lattice will change and the individual grains will adjust their shape. The mutual effects of grains bring about additional elastic deformation in each grain and thus also internal stresses  $\sigma$ .

The resulting elastic deformation is difficult to determine. According to [5], poling can cause such deformations in ceramics with tetragonal structure that the mean value would amount to  $0.368e_{T3}$  in an ideal case. It may therefore be assumed that even elastic deformation, which varies with the individual grains, can attain the maximum values of elongation and contraction of that size. At a number of points of the polycrystalline material there thus arise elastic elongations  $\varepsilon$  perpendicularly to the grain boundaries; on the basis of data given in [5], the elongation should amount to  $\varepsilon \approx 1/3e_{T_3}$ , i.e.  $\varepsilon \doteq 6 \times 10^{-3}$ .

According to the isotropic theory of elasticity, the maximum tensile stress acting perpendicularly to the grain boundaries is given by the equation

$$\sigma = E\varepsilon, \qquad (1)$$

where E is Young's modulus of elasticity.

Using Young's modulus of elasticity for piezoceramics,  $E \doteq 80$  GPa [6] and  $\varepsilon \doteq 6 \times 10^{-3}$ , one obtains  $\sigma \doteq 480$  MPa for maximum tensile stresses perpendicular to the grain boundaries. The value of these internal stresses does not depend on grain size. Under certain conditions the internal stresses can cause cracking along the grain boundaries. However, the actual process by which, the cracks are formed, depends on grain size d and it appears that there exists a critical grain size  $d_c$  such that cracks will form at grain sizes  $d > d_c$  as a result of poling which will cause no cracking at grain sizes  $d < d_c$ .

To determine  $d_c$ , it is possible to use suitably modified relationships that have been derived for the formation of cracks in polycrystalline materials due to anisotropy of thermal expansion [7, 8, 9, 10].

Let us consider cube-shaped grains with edges d in length. If tensile stress  $\sigma$  acts in a certain grain, the grain will exhibit the following density of elastic energy:

$$w = \frac{\sigma^2}{2E}.$$
 (2)

The total elastic energy in the grain is thus

$$W = \frac{\sigma^2}{2E} d^3. \tag{3}$$

A part of this energy,  $\Delta W$ , is released when a crack forms along the boundary with the neighbouring grain, in the direction perpendicular to the tensile stress. It therefore holds that  $\Delta W = kW$ , where 0 < k < 1.

Energy  $\gamma_1$  is required for creating a unit area of crack along the grain boundary  $(\gamma_1 \text{ is the energy of intercrystalline fracture})$ . The energy necessary for producing a crack of area  $d^2$  between two grains is

$$W_1 = \gamma_1 d^2. \tag{4}$$

The crack can form along the grain boundary only when the loss in the elastic energy of the grain,  $\Delta W$ , is capable of supplying energy  $W_1$  required for the crack formation, i.e. when  $\Delta W > W_1$ . From the condition  $\Delta W = W_1$  it is possible to calculate the critical grain size value,  $d_c$ :

$$d_c = \frac{2}{k} \frac{\gamma_1 E}{\sigma^2} \,. \tag{5}$$

The ratio of the relaxed energy,  $\Delta W$ , to the total elastic energy of the grain W can be estimated as  $k \approx 1/5$ .

Using equation (1), one obtains

$$d_c = K \frac{\gamma_1}{E\varepsilon^2} \tag{6}$$

where  $K = \frac{2}{k} \approx 10$ .

The derivation of equation (6) involves a number of simplifying assumptions. Primarily, it is based only on energy considerations and assumes that creation of the crack is possible if adequate energy for the final state is available. Actually, there must be a nucleus of the crack, from which it can propagate if adequate concentrated stresses arise at the crack front. However, ceramics made by sintering always contain a number of pores at the grain boundaries as well as microcracks at the points of contact of three grains, so that there are actually conditions for crack propagation over the entire grain boundary area, if  $d > d_c$ . If, however, the grain boundaries are perfectly bound, no crack need form even at  $d > d_c$ , as the crack nucleation is generally a process much less readily occurring than crack propagation.

The assumption of a cube-shaped form of the grains was another simplification. The grain boundary facets have in fact areas smaller than  $d^2$ . None the less, the detailed theories employed in the derivation of  $d_c$  for anisotropy of thermal expansion [7, 8, 9, 10] also lead to a relationship similar to equation (6) where the constant K has values between 10 and 15 [7].

The value of fracture energy along the grain boundaries is difficult to determine. However, it can be assumed to lie between the minimum possible value  $\gamma_{1\min} = 2\gamma_0 - \gamma_{GB}$  (where  $\gamma_0$  is surface energy and  $\gamma_{GB}$  is the energy of the grain boundary), and the maximum possible value  $\gamma_{1\max} = \gamma_{ef}$ , where  $\gamma_{ef}$  is the effective fracture energy of a macroscopic specimen where, of course, the crack encounters additional obstacles when passing from one grain to another.

For piezoceramics of the type PbTiO<sub>3</sub>-PbZrO<sub>3</sub>, the effective fracture energy,  $\gamma_{ef}$ , found on macroscopic specimens with a mean grain size  $\bar{d} = 5 \,\mu$ m, was about  $5 \,\text{J/m}^2$  [6]. Let us take the probable value of fracture energy along the grain boundaries  $\gamma_1 = \frac{1}{2} \gamma_{ef} \approx 2.5 \,\text{J/m}^2$ . On taking the constant K in equation (6) as being equal to 12, as the most probable value, and substituting the given values of  $\varepsilon$ ,  $E, \gamma_1$  and K into equation (6), one obtains an estimate of the critical size  $d_c \doteq 10 \,\mu$ m. This estimate shows a relatively low accuracy, above all in view of the inaccuracy in the determination of  $\gamma_1$  and  $\varepsilon$ .

However, the theoretical estimate makes it possible to predict that poling of the given fine-grained piezoceramics with a mean grain size of  $\bar{d} = 6 \,\mu\text{m}$  will not lead to cracking. Large number of cracks will form in a coarse-grained material with a mean grain size  $\bar{d} = 16 \,\mu\text{m}$ , particularly along the boundaries of grains larger than the mean grain size.

#### **RESULTS AND THEIR EVALUATION**

According to Fig. 2, the bending strength of the fine-grained ceramics  $(\bar{d} = 6 \,\mu\text{m})$  is  $R_0 = 135 \pm 25$  MPa in the non-poled state. The value remains virtually unchanged or shows a mild increase as a result of poling over the entire intensity range of the DC electrical field. The fractographic examination (Fig. 3a, b, c) shows a roughly identical pattern of fracture in the poled and non-poled state, the shares of intercrystalline and transcrystalline fracture being approximately the same. This agrees with the conclusion that no cracking along grain boundaries occurs in the fine-grained specimens as a result of poling.



Fig. 2. Bending strength R of the transducers in dependence on poling electric field: a) fine-grained ceramics, b) coarse-grained ceramics.

The bending strength of the coarse-grained material  $(d = 16 \ \mu\text{m})$  in nonpoled state was  $R_0 = 90 \pm 10$  MPa, i.e. lower than of the fine-grained material. This corresponds to the well known indirect dependence of strength on grain size in ceramics (e.g. [7]). The fracture surface of the non-poled specimen (Fig. 4a) shows also transcrystalline fracture apart from intercrystalline fracture. Poling in an electrical field of 1 kV/mm intensity virtually does not change bending strength (Fig. 2) nor the character of fracture (Fig. 4b), which conforms to the theoretical conception of the movement of the 180-degree domain walls, which does not bring about creation of internal stresses. (For Figs. 3—6 see Appendix.)

The transducers poled in a 3.2 kV/mm field showed a low bending strength and the fracture surface exhibited only intercrystalline fracture (Fig. 4c). According to the analysis given above, this corresponds to the effect of internal stresses arising during movement of the 90-degree walls and causing formation of cracks along the grain boundaries. In such a material, the crack propagation proceeds readily through gradual joining of cracks formed in the course of poling. The analysis also conforms to the gradual decrease of bending strength with increasing intensity of the poling field. The state attained at such field intensities which already cause the 90-degree domains to reorient but do not yet bring about the saturated mean deformation  $\varepsilon \doteq (1/3)e_{T_s}$  corresponds to the larger critical dimension  $d_c$  according to equation (6). In specimens with a certain grain size distribution, the cracks arise only at the boundaries of the larger grains. The density of the cracks is therefore lower, and the bending strength higher than with the material which has been poled to the saturated state.

The cracks arising due to poling of coarse-grained piezoceramic materials were well visible on polished sections under an optical microscope. Fig. 5a shows a micrograph of a non-poled fine-grained specimen. The micrographs of the material poled in electric fields of 1.0 kV/mm and 3.2 kV/mm intensity respectively are shown in Fig. 5b, c. There is no discernible difference between the micrographs. The analogous micrographs of specimens of coarse-grained ceramics are shown in Fig. 6a, b, c.

The polished sections of the non-poled material and that poled in the 1 kV/mm field have the same character, being free of discernible cracks. The micrograph of the material poled in the 3.2 kV/mm field already shows well visible cracks due to poling (Fig. 6c).

The presence of cracks affects significantly the oscillations of poled coarse-grained transducers. Fig. 7 shows the factors of mechanical quality in terms of the intensity of the poling field. The diagram indicates that the mechanical quality factor of coarse-grained piezoceramics decreases strongly with the formation of cracks during poling. With coarse-grained materials poled at electric field intensities higher than 2 kV/mm the  $Q_{\text{M}}$  values were so low that they could not be determined from characteristic frequency measurements.

Oscillation of transducers containing cracks involves mild opening of the cracks, absorption of energy at the points of stress concentration in the crack fronts, friction of the crack areas and dispersion of elastic waves at the cracks. All these factors contribute to damping the elastic waves and thus to decreasing  $Q_{21}$ .

A decrease of the  $Q_M$  values with increasing poling electric field was observed by Yamashita [11] on transducers of the PbTiO<sub>3</sub> ceramics, and by Wersing [12] on piezoceramics based on Pb ( $Mg_{1/2} W_{1/2}$ ) TiZrO<sub>3</sub> solid solutions. These authors did not compare the properties of fine- and coarse-grained ceramics in the given systems and did not determine the mechanical properties. Wersing [12] attributes the low  $Q_M$  values to viscoelastic phenomena at the grain boundaries of the poled transducers. We suggest that the formation of cracks due to poling of coarsegrained ceramics of the system Pb( $Mg_{1/3}Nb_{2/3}$ )TiZrO<sub>3</sub> provides a satisfactory explanation of the low  $Q_M$  values observed.

## CONCLUSION

Poling of piezoelectric transducers brings about reorientation of their domain structure. If the intensity of the electric field is so high that the 90-degree domain walls move in a ceramic material with a tetragonal structure, internal stresses will arise in the transducers and lead to the formation of cracks if a certain critical grain size of the material has been exceeded.

Piezoelectric transducers of the solid solution  $Pb(Mg_{1/3}Nb_{2/3})_{0.44}Ti_{0.44}Zr_{0.12}O_3$ , doped with 1 wt% MnO<sub>2</sub>, were prepared in fine-grained and coarse-grained forms by changing the initial PbO content. The critical grain size has been exceeded with the coarse-grained materials. Their poling resulted in the formation of cracks which in turn reduced the bending strength, and the mechanical quality factor of the oscillating transducers.

From the derived equation for the critical grain size it follows that this size decreases with increasing distortion of the ferroelectric phase in relation to the high-temperature paraelectric phase. With coarse-grained piezoceramics based on Pb(Ti,  $ZrO_3$  solid solutions, cracking due to poling can therefore be expected particularly in the case of PbTiO<sub>3</sub> ceramics or those with a tetragonal structure and an elevated content of lead titanate.

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# VLIV VELIKOSTI ZRN NA VLASTNOSTI PIEZOKERAMIKY ZE SYSTÉMU Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>-PbTiO<sub>3</sub>-PbZrO<sub>3</sub>

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Piezoelektrické měniče z pevného roztoku Pb $(Mg_{1/3}Nb_{2/3})_{0,44}Ti_{0,44}Zr_{0,12}O_3$  (obr. 1) dopovaného 1% hmotn. MnO<sub>2</sub> bylo možno změnou obsahu PbO ve výchozí směsi surovin připravit jednak v jemnozrnném, jednak v hrubozrnném stavu. Jemnozrnná keramika (obr. 3) o střední velikosti zrn 6 µm byla připravena ze směsi oxidů odpovídajících stechiometrickému složení pevného roztoku. Hrubozrnná keramika o střední velikosti zrn 16 µm byla získána ze směsí oxidů, ve které byl obsah PbO zvýšen o 3% hmotn. (obr. 4). Připravené měniče mají tetragonální strukturu.

Při polarizaci těchto měničů dochází k pohybu stoosmdesátistupňových a devadesátistupňových doménových stěn. Pohyb devádesátistupňových stěn je spojen se vznikem vnitřních pnutí. Tato pnutí mohou vést ke vzniku trhlin, jestliže rozměry zrn piezokeramického měniče překračují určitou kritickou hodnotu (rov. (6)). Výraz pro kritický rozměr zrn byl získán modifikací vztahů odvozených pro kritický rozměr zrn keramického materiálu, v němž vznikají trhliny vlivem anizotropie teplotní roztažnosti [7, 8, 9, 10].

U připravených hrubozrnných měničů velikosti zrn tento kritický rozměr překračují. Při polarizaci elektrickým polem vysoké intenzity, které vyvolává posuny devadesátistupňových doménových stěn, ke vzniku trhlin skutečně dochází. Tyto trhliny bylo možno pozorovat v optickém mikroskopu na leštěných nábrusech polarizovaných měničů (obr. 6c).

Trhliny vzniklé při polarizaci hrubozrnných měničů jsou příčinou zhoršení pevnosti v ohybu (obr. 2). U kmitajících měničů přítomnost trhlin zvyšuje tlumení elastických vln, což se projevuje snížením činitele mechanické jakosti (obr. 7).



Fig. 7. Mechanical quality factor  $Q_M$  vs. the intensity of electric field during poling; a) fine-grained ceramics, b) coarse-grained ceramics.

- Obr. 1. Stavový diagram systému Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>—PbTiO<sub>3</sub>—PbZrO<sub>3</sub>: T tetragonální fáze, R — romboedrická fáze, PC — pseudokubická fáze. Složení pevného roztoku připravených měničů je vyznačeno bodem F.
- Obr. 2. Závislost pevnosti v ohybu R měničů na intenzitě elektrického pole při polarizaci; a) jemnozrnná, b) hrubozrnná keramika.
- Obr. 3. Lomová plocha měniče z jemnozrnné keramiky;
  a) nepolarizovaný měnič, b) měnič polarizován el. polem 1 kV/mm, c) měnič polarizován el. polem 3,2 kV/mm.
- Obr. 4. Lomová plocha měniče z hrubozrnné keramiky; a) nepolarizovaný měnič, b) měnič polarizován el. polem 1 kV/mm, c) měnič polarizován el. polem 3,2 kV/mm.
- Obr. 5. Leštěný nábrus měniče z jemnozrnné keramiky;
  - a) nepolarizovaný měnič, b) měnič polarizován el. polem 1 kV/mm, c) měnič polarizován el. polem 3,2 kV/mm.
- Obr. 6. Leštěný nábrus měniče z hrubozrnné keramiky;
   a) nepolarizovaný měnič, b) měnič polarizován el. polem 1 kV/mm, c) měnič polarizován el. polem 3,2 kV/mm.
- Obr. 7. Závislost činitele mechanické jakosti měničů na intenzitě el. pole při polarizaci: a) jemnozrnná, b) hrbozrnná keramika.

ВЛИЯНИЕ РАЗМЕРА ЗЕРЕН НА СВОЙСТВА ПИЕЗОКЕРАМИКИ, ПОЛУЧЕННОЙ ИЗ Рb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>—PbTiO<sub>3</sub>—PbZrO<sub>3</sub>

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Писзоэлектрические обменники из твердого раствора Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)<sub>0,44</sub>Ti<sub>0,44</sub>Zr<sub>0,12</sub>●<sub>3</sub> (рис. 1) с добавкой 1 % по весу MnO<sub>2</sub> можно изменением содержания PbO в исходной смеси сырья получать как в тонкозернистом, так и грубозернистом виде.

Тонкозернистую керамику (рис. 3) средним размером зерна 6 µм получали из смеси оксидов, отвечающих стехиометрическому составу твердого раствора. Грубозернистую керамику средним размером зерна 16 µм получали из смеси оксидов, в которой содержание РьФ повышали на 3 % по весу (рис. 4). Полученные обменники имеют тетрагональную структуру.

При поляризации приводимых обменников происходит движение 180-градусных и 90-градусных доменных стен. Движение 90-градусных стен связано с образованием внутренних напряжений. Данные напряжения могут вызывать образование трещин в том случае, когда размеры зерен пиезокерамического обменника превышают определенный критический предел (уравнение 6). Выражение для критического размера зерен получали модификацией отношений, выведенных для критического размера зерен керамического материала, в котором образуются трепины под влиянием анизотропии теплового расширения [7, 8, 9, 10].

У полученных грубозернистых обменников размеры зерен данный критический размер превышают. При поляризации электрическим полем высокой интенсивности, которое вызывает смещения 90-градусных доменных стен, образование трещин действительно происходит. Приводимые трещины можно наблюдать с помощью микроскопа на отполированных аншлифах поляризованных обменников (рис. 6с).

Трещины, образовавшиеся при поляризации грубозернистых обменников являются причиной понижения прочности при изгибе (рис. 2). У колеблющихся обменников присутствие трещин повышает гашение эластических волн, что проявляется в понижении коэффициента механического качества (рис. 7).

- Рис. 1. Диаграмма состояния системы Pb(Mg<sub>1/3</sub>Nb<sub>2/3</sub>)O<sub>3</sub>—PbTiO<sub>3</sub>—PbZrO<sub>3</sub>: T тетрагональная фаза, R — ромбоэдрическая фаза, PC — псевдокубическая фаза. Состав твердого раствора полученных обменников обозначен через точку F.
- Рис. 2. Зависимость прочности при изгибе R обменников от интенсивности электрического поля при поляризации; а) тонкозернистая керамика, b) грубозернистая керамика.
- Рис. 3. Поверхность излома обменника из тонковернистой керамики; а) неполяризованный обменник, b) обменник, поляризованный электрическим полем 1 кв/мм, с) обменник, поляризованный электрическим полем 3,2 кв/мм.
- Рис. 4. Поверхность излома обменника из грубозернистой керамики; а) неполяризованный обменник, ь) обменник, поляризованный электрическим полем 1 ко/мм, с) обменник, поляризованный электрическим полем 3,2 кв/мм.
- Рис. 5. Отполированный аншлиф обменника из тонкозренистой керамики: а) неполяризованный обменник, b) обменник, поляризованный электрическим полем 1 кв/мм, c) обменник, поляризованный электрическим полем 3,2 кв/мм.
- Рис. 6. Отполированный аншлиф обменника из грубозернистой керамики; а) неполяризованный обменник, b) обменник, поляризованный электрическим полем I кв/мм, c) обменник, поляризованный электрическим полем 3,2 кв/мм.
- Рис. 7. Зависимость коэффициента механического качества обменников от инпенсивности электрического поля при поляризации; а) тонковернистая керамика, b) грубовернистая керамика.



3a

3Ъ



Fig. 3. Fracture surface of the fine-grained transducer; a) non-poled transducer, b) transducer poled in a 1 kV/mm field, c) transducer poled in a 3.2 kV/mm field.



Fig. 3c

4a



Fig. 4. Fracture surface of the coarse-grained piezoceramics; a) non-poled transducer, b) transducer poled in a 1 kV/mm field, c) transducer poled in a 3.2 kV/mm field.



Fig. 4b







5a

5b



Fig. 5. Polished section of the fine-grained ceramic transducer; a) non-poled transducer b) transducer poled in a 1 kV/mm field, c) transducer poled in a 3.2 kV/mm field.

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Fig. 5c



Fig. 6. Polished section of the coarse-grained ceramic transducer: a) non-poled transducer, b) tranducer poled in a 1 kV/mm field, c) transducer poled in a 3.2 kV/mm field.

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6a



Fig.6b

 $Fig. \ 6c$ 

