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Microwave absorption properties of ferroelectric piezoceramic materials

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The results of a study in the microwave range of the microwave absorbing properties of piezoceramic materials developed at the Scientific Research Institute of Physics of the Southern Federal University and based mainly on the $PbTiO_3-PbZrO_3$ system are presented in comparison with industrial composite materials based on carbonyl iron and epoxy resin and materials of industrial dielectric resonators. A technique for measuring and calculating the parameters of samples of the materials under study is described. The relationship between the microwave absorbing properties of the ceramics under study and their phase composition has been established. An assessment has been made of the applicability of the developed piezoceramic materials in microwave technology devices

Keywords: ferroelectrics, piezoelectric, microwave absorption, microstrip line.

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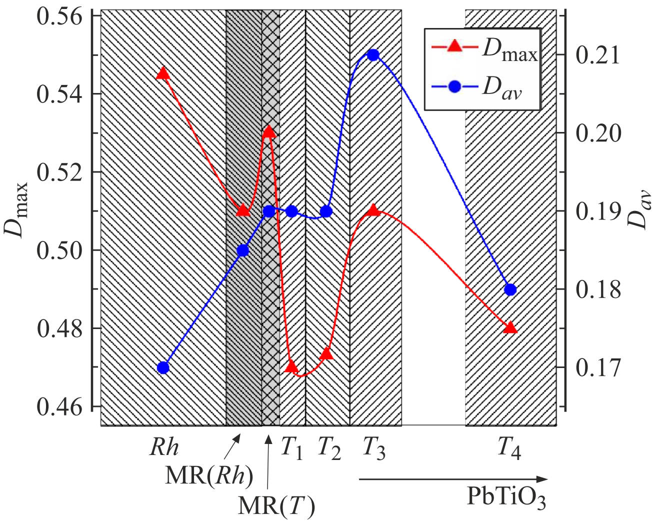
Solid solutions of multicomponent systems of the $PbTiO_3-PbZrO_3-\sum_{i=0}^{n}(PbB'_{1-\alpha}B''_{\alpha}O_3)_n$ type with lead being their key element form the basis of almost all ferroelectric piezoceramic materials (FEPCMs) developed around the world. Their noteworthy piezoelectric properties depend largely on their polarization properties [1].

FEPCMs are used widely in MEMS (microelectromechanical systems) [2,3], which may serve as components of microwave devices [4]. Without proper shielding, they, however, may exert a negative effect on the purity of the microwave signal spectrum. At the same time, certain properties of FEPCMs, such as their high permittivity in the microwave range and fairly high piezocoefficients, may be an advantage in the development of reflector antennas or voltage-tunable resonators [5,6]. In view of the above, special attention is paid to the study of losses in microwave devices manufactured with the use of piezoceramics [7].

The aim of the present study is to investigate the specifics of formation of the resonant and non-resonant microwave absorption level in FEPCMs with different phase filling developed at the Scientific Research Institute of Physics of the Southern Federal University (SFedU).

PCR-type (Rostov piezoceramics) industrial piezoceramic materials of various applications listed in Table 1 were examined [1,8]. FEPCMs were fabricated by solid-phase synthesis with subsequent sintering in accordance with the conventional ceramic technology [9]. Samples of all materials had the form of cylinders with a diameter of 10.0 ± 0.1 mm and a height of 1.00 ± 0.03 mm. Radiophysical measurements were performed using a Keysight P9375A vector network analyzer (United States) with an operating frequency range of 300 kHz–26.5 GHz and a measuring cell of the following design: a straight section of a microstrip line (MSL) placed on a substrate made of an epoxy material reinforced with fiberglass (FR4) with SMA 3.5 mm coaxial connectors at both ends. The measurements were carried out in two configurations (with samples positioned on the straight MSL section and next to the MSL on the substrate). The sample positioning near the MSL was identical to that found in the design of a generator based on a dielectric resonator with series feedback [10] and was used to establish the resonant nature of loss maxima. The procedure outlined in [11,12] was used to calculate the



Dependence of absorption parameters of multicomponent PZT-based solid solutions on the $PbTiO_3$ concentration. MR — morphotropic region, *Rh* — rhombohedral phase, and *T* — tetragonal phase.

Table 1. FEPCMs of various applications developed at the Scientific Research Institute of Physics of SFedU

Group	Field of application	Materials
1 (materials resistant to electric and mechanical influence)	Power devices (piezoelectric transformers, piezoelectric motors, ultrasonic radiators, and high-voltage generators)	PCR-12, PCR-22 PCR-23, PCR-6 PCR-77, PCR-78, PCR-8, PCR-86
2 (materials with high permittivity values)	Low-frequency receiving equipment (hydrophones, microphones, and seismic detectors)	PCR-66, PCR-7, PCR-7M
3 (materials with a high sensitivity to mechanical stress)	Accelerometers, ultrasonic analyzers, devices for nondestructive materials testing by acoustic emission, and instruments for ultrasonic diagnostics in medicine	PCR-37
4 (materials with moderate permittivity values)	Transducers operating in the receiving mode with a high sensitivity both under load (specific sensitivity), and no-load conditions (materials with high values of $d_{ij}/\sqrt{\varepsilon_{33}^T/\varepsilon_0}$ and g_{ij})	PCR-88, PCR-89
5 (materials with a strong anisotropy of piezoelectric parameters)	Ultrasonic analyzers, medical diagnostic instruments, accelerometers, and piezoceramic sensors with an elevated sensitivity to hydrostatic pressure	PCR-40
6 (materials with a high stability of the resonant frequency of electromechanical oscillations)	Filter devices	PCR-13, PCR-74
7, 8 (pyroelectric materials and materials with a low permittivity)	High-frequency acoustoelectric transducers and sensors of pyroelectric thermal energy detectors	PCR-10, PCR-11, PCR-3

coefficient of scattering of electromagnetic wave energy by the sample (radiation losses and ohmic losses in the sample included) in all measurement configurations for each measured frequency. The key parameters chosen to characterize microwave absorption in the materials under study were the most pronounced maximum of the scattering coefficient $\big((D_i)_{\max} = D_i(f_{\max})$: $\forall f \in [f_{ini}, f_{fin}]$, $f \neq f_{\max}$: $D_i(f) \leqslant D_i(f_{\max})$, f_{ini} and f_{fin} are the lower and upper boundaries of the frequency measurement range$\big)$, its frequency ($f_{\max}$, GHz), and the average level of scattering ($(D_i)_{av}$) within the entire frequency measurement range (300 kHz–26.5 GHz), which was calculated as

$$(D_i)_{av} = \frac{1}{f_{fin} - f_{ini}} \int\limits_{f_{ini}}^{f_{fin}} D_i(f)df.$$

In the case of detection of resonance maxima of samples located near the MSL, the lowest-frequency maximum was approximated by a Gaussian function, and its loaded quality factor was estimated as $Q_i = \frac{f_{\max}}{\Delta f}$,

Table 2. Microwave absorption parameters of PZT-based FEPCMs

Material	Symmetry	$D_{\max}$	D_{av}
Region Rh			
Group 8, PCR-10	T	0.47	0.19
Group 8, PCR-11	Rh	0.54	0.17
Group 8, PCR-3	Rh	0.55	0.17
Morphotropic region near Rh			
Group 1, PCR-23	$T + Rh$ (traces)	0.5	0.19
Group 3, PCR-37	$60T + 40Rh$	0.48	0.17
Group 6, PCR-74	$55T + 45Rh$	0.52	0.18
Morphotropic region near T			
Group 2, PCR-7	T	0.53	0.19
Group 2, PCR-7M	$75T + 25Rh$	0.53	0.19
Group 2, PCR-66	$80T + 20Rh$	0.5	0.19
Group 4, PCR-88	$T + Rh$	0.5	0.19
Group 4, PCR-89	$80T + 20Rh$	0.46	0.19
Region T_1			
Group 1, PCR-77	$90T + 10Rh(PSC)$	0.5	0.18
Group 1, PCR-78	$90T + 10Rh$	0.49	0.19
Group 1, PCR-8	$85T + 15Rh$	0.45	0.19
Region T_2			
Group 1, PCR-12	T	0.49	0.19
Group 1, PCR-22	T	0.46	0.19
Group 1, PCR-6	$T + PSC$(traces)	0.47	0.19
Group 1, PCR-86	$T + Rh + PSC$	0.51	0.19
Region T_3			
Group 6, PCR-13	T	0.51	0.21
Region T_4			
Group 5, PCR-40	T	0.48	0.18

where Δf is the width of the maximum at half-power level [13]. The permittivity of materials and the dielectric loss tangent at low frequencies (1 kHz) were investigated using an Agilent E4980A *LCR* meter (United States). X-ray studies were performed using the powder diffraction method with a DRON-3 diffractometer (CoK_α radiation) connected to a PC with specialized software installed on it. Structural parameters were calculated in accordance with standard procedures [14].

Most of the studied FEPCMs belong to different regions of phase diagrams (PDs) of solid solutions of multicomponent systems based on $PbTiO_3-PbZrO_3$ (PZT) presented in [1]. Table 2 lists the parameters characterizing the absorption level of the examined materials, their corresponding PD regions, and the actual phase compositions of samples determined by analyzing X-ray data (Rh — rhombohedral symmetry, T — tetragonal symmetry, and PSC — pseudocubic symmetry).

Having calculated the average values of the presented absorption parameters for each PD region, we obtained the dependence of these parameters of the studied solid solutions on the $PbTiO_3$ concentration (see the figure).

High maximum values of the scattering coefficient in the microwave range and its comparatively low average level are attributable to resonant absorption of energy in the samples, which arises from low permittivity values typical of solid solutions of the PZT system in the Rh phase. When one approaches the morphotropic region from the direction of phase Rh, the maximum level of the scattering coefficient becomes lower due to the suppression of influence of the resonant response in samples with a shift of the resonant maxima to the high-frequency region. This is related to the growth of permittivity, which, in turn, is caused by an increase in mobility of domain walls in ferroelectric soft materials. However, in the morphotropic region, the level of losses increases locally on the T phase side due to structural instabilities in materials and the breaking of chemical bonds in the process of formation of a new crystalline phase. As one enters the T region and moves further along the PD towards increasing $PbTiO_3$ concentration, the maximum level of losses increases slightly, while the average level of losses decreases. This is due to a reduction in permittivity and dielectric losses in materials, which, in turn, is attributable to an enhancement of their ferroelectric hardness [1]. The high maximum and average values of losses in PCR-13 from the T_3 PD region are worth noting. These losses may be attributed to the heterogeneity of solid solutions in this region due to the formation of intermediate fields of transition from one T phase to another and the resulting structural instabilities [15].

PCR-13 has the highest level of losses among all the studied FEPCMs. Losses in the indicated composition are comparable to those in industrial absorbers based on 60% carbonyl iron, while the absorption band is positioned at lower frequencies.

The following conclusions may be drawn based on the obtained data: the highest level of losses is found in FEPCMs belonging to the morphotropic PD region; PCR-13 has the highest level of losses among all the studied FEPCMs (losses in it are comparable to those in an industrial absorber); therefore, this material is a potential candidate for fabrication of microwave-absorbing coatings operating within the 3.0–8.0 GHz range.

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Conflict of interest

The authors declare that they have no conflict of interest.

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