

Influence of titanium oxide addition on properties of lead zirconate titanate-nickel ferrite ceramics

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The influence of titanium oxide additions on magnetoelectric effect in bulk PZT850 — nickel ferrite composites has been studied. It has been found that the bulk composite doping with titanium oxide in an amount up to 6 % increases the magnetoelectric voltage coefficient by 30–40 %, magnetoelectric polarization by 80–150 %, dielectric constant by 45–80 %. These results are explained by formation of a phase interlayer enriched in titanium oxide, which prevents the mutual solubility of the phases.

Исследовано влияние добавок оксида титана на магнитоэлектрический эффект в объемных композиционных материалах PZT850 — феррит никеля. Установлено, что легирование объемных композиционных материалов окисью титана в количестве до 6 % увеличивает магнитоэлектрический коэффициент по напряжению на 30–40 %, магнитоэлектрический коэффициент по поляризации на 80–150 %, диэлектрическую проницаемость на 45–80 %. Полученный результат объясняется образованием межфазной прослойки, обогащенной оксидом титана, которая препятствует взаимному растворению фаз.

1. Introduction

The sintering of bulk ferrite — piezoelectric composites is accompanied by mutual dissolution of phases and cross-doping of initial components resulting in reduced magnetoelectric (ME) effect [1, 2]. It is known that introduction of additions allows to influence the ceramics sintering and to improve its properties. This problem was not considered with due attention in the field of bulk composite preparation [3]. Therefore, it is of interest to study properties of the bulk composites obtained using various additions.

2. Experiment

This paper presents the research results on titanium oxide addition influence on the ME characteristics of bulk composites in the lead zirconate titanate — nickel ferrite system. The material used was the composite ceramics based on the industrial piezoceramics PZT850 and modified nickel ferrite $NiFe_{1.9}Co_{0.02}O_4$ with the ferrite to piezoelectric ratio 25:75 (Composition 1) and 50:50 (Composition 2). The amount of TiO_2 additions was 0.5, 1, 2, 4, 6 and 8 wt.% with respect to ferrite with piezoelectric. The ME effect was studied by measuring the alternating voltage that appeared in the sample under the simultaneous influence of variable and constant magnetic fields. The ME voltage coefficient dE/dH was determined proceeding from the sample thickness h, voltage dV and the variable magnetic field intensity dH, $dE/dH = dV/(h \cdot dH)$. The investigations have been carried out for two different sample orientations at 1 kHz frequency. In one case, the electric polarization vector was perpendicular to magnetic

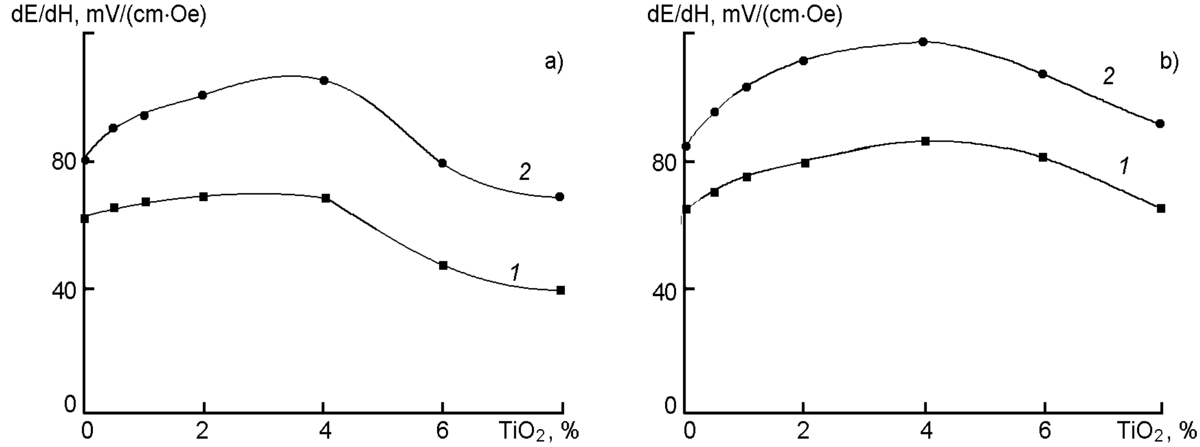

Fig. 1. ME voltage coefficient dependences on TiO_2 content for bulk composites of Compositions 1 (a) and 2 (b): curve *1* – transverse; *2* – longitudinal.

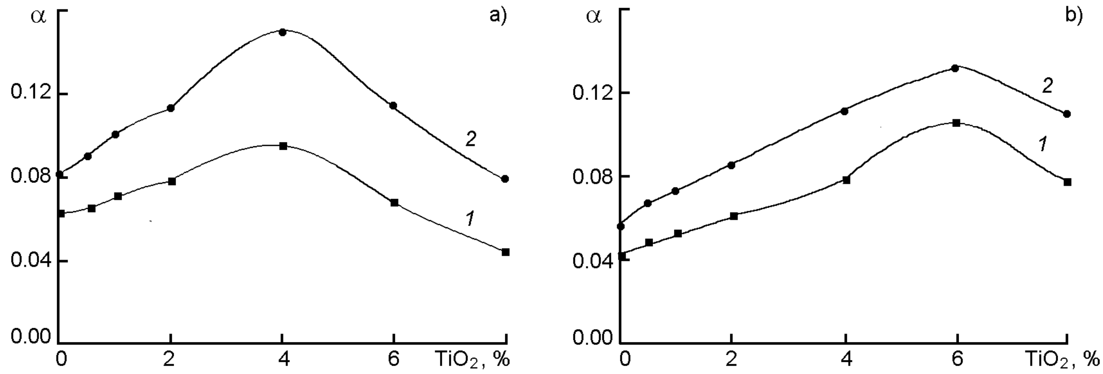

Fig. 2. ME polarization coefficient dependences on TiO_2 content for bulk composites of Compositions 1 (a) and 2 (b): curve *1* – transverse; *2* – longitudinal.

fields (transverse effect), while in other case, parallel thereto (longitudinal effect). Basing on the data obtained, the values of the ME polarization coefficient $\alpha = 4\pi dP/dH$ have been calculated.

3. Discussion

The study results on the influence of titanium oxide additions on the ME voltage coefficient are presented in Fig. 1. The diagram shows that the titanium oxide introduction additions in the first composition increases the mentioned coefficient by 30 %. For the second composition, this parameter increases by 40 %. As to the ME polarization coefficient (Fig. 2), the titanium oxide addition increase that coefficient for the first composition up to 80 %, for the second one, by a factor of about 2.5. To understand the mechanism of this phenomenon, we consider the effect of titanium oxide on the dielectric properties of the studied materials.

One of necessary conditions for the existence ME effect is the material polarization. The ME effect depends on the value of polarization. The existence of the magnetic phase reduces the composite resistivity as compared to the pure piezoelectric material, thus worsening the polarization conditions. Therefore, we used the modified nickel ferrite ($NiFe_{1.9}Co_{0.02}O_4$) having a high resistivity. In addition, the titanium oxide introduction increases the composite resistivity by 2 to 3 times. This results in a reduction of the leakage currents during the polarization and, as a consequence, in enhanced piezoelectric and ME characteristics.

The investigation of dielectric properties has shown that the introduction of titanium oxide additions causes an increase of dielectric permeability for the first composition by 50 %, while for the second, by 70 %. It is also accompanied by a halved dielectric loss tangent (Fig. 3). Let the possible causes of this phenomenon be considered. The composite material consists of two phases: mag-

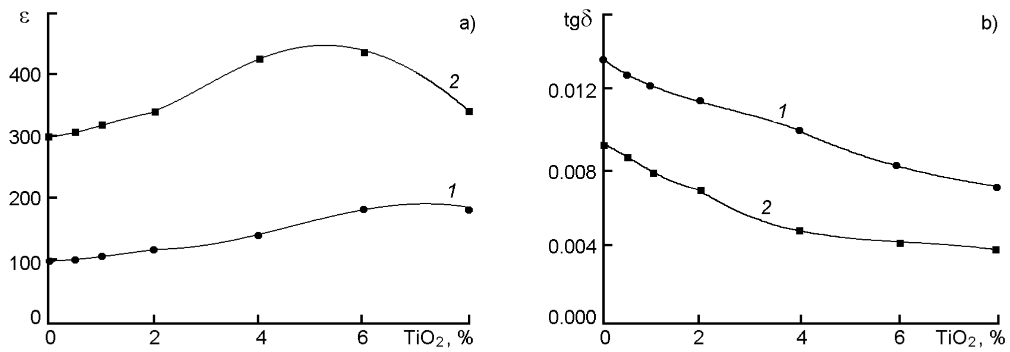

Fig. 3. Dependences of dielectric permeability (a) and dielectric loss tangent (b) on TiO_2 content for bulk composites of Compositions 1 (curve *1*) and 2 (curve *2*).

netic and piezoelectric. It is well-known that dielectric permeability of ferrite can reach tens and hundreds of thousands. However, it is always accompanied by increase of dielectric loss tangent. In our case, the dielectric loss tangent decreases. Hence, the magnetic phase cannot be responsible for this phenomenon. The studies on the influence of titanium oxide additions on the PZT850 dielectric properties have not revealed any increase in the piezoceramics dielectric permeability. Therefore, that the obtained result can be assumed to be a consequence of the two-phase structure presence in the investigated materials and explained as follows.

An interphase layer is formed during the bulk composite sintering. The titanium oxide additions increase its resistance and prevent the mutual dissolution of phases. The consequence is the increase in specific resistance, dielectric permeability and reduction of dielectric loss tangent of the composites. These parameters affect directly the ME coefficents, promoting their growth. The investigation of the sintering temperature influence on the dielectric permeability has confirmed this mechanism. The dielectric permeability of the investigated materials decreases with the sintering temperature elevation. In the composites modified with titanium oxide, this dependence is less pronounced as compared to the initial materials. Similar results were obtained in a study of ceramic composite materials for the barium titanate — nickel ferrite system. Such regularities can be assumed to be relevant to all bulk composites on the basis of perovskite and spinel materials.

4. Conclusions

The influence of titanium oxide addition on the ME effect in bulk composites has shown an increase of the ME voltage coefficient by 30–40%. For ME polarization coefficient, this increase was from 80 to 150 %. The obtained results are explained by the formation of an interphase layer enriched in titanium oxide, which prevents the mutual dissolution of the phases.

References

1. J.Ryu, A.Vazqez, K.Uchino et al., *J.Electroceramics*, **7**, 17 (2001).
2. J.Y.Zhai, N.Cai, L.Liu et al., *Mater. Science and Engin.*, **B99**, 329 (2003).
3. J.van den Boomgard, R.A.J.Born, *J.Mater. Science*, **13**, 1538 (1978).

Вплив домішок оксиду титану на властивості кераміки цирконат-титанат свинцю-ферит нікелю

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Досліджено вплив домішок оксиду титану на магнітоелектричний ефект в об'ємних композиційних матеріалах PZT850 — ферит нікелю. Встановлено, що легування об'ємних композиційних матеріалів оксидом титану в кількості до 6 % збільшує магнітоелектричний коефіцієнт за напругою на 30–40 %, магнітоелектричний коефіцієнт за поляризацією на 80–150 %, діелектричну проникність – на 45–80 %. Одержаний результат пояснюється утворенням міжфазового прошарку, збагаченого оксидом титану, який перешкоджає взаємному розчиненню фаз.