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$Y_3Fe_5O_{12}$/Na,Bi,Sr-doped PZT particulate magnetoelectric composites

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ABSTRACT

Magnetoelectric (ME) composites of Na, Bi, Sr substituted lead zirconate titanate (PZT) and yttrium iron garnet having representative formula $(100-x)$ wt% Na,Bi,Sr-doped PZT (PZTNB-1)$+x$ wt% $Y_3Fe_5O_{12}$ (YIG) with $x=10$–90 were manufactured using powdered components obtained through sol–gel processes. It is shown that the decrease in sintering temperature provided by the use of finely dispersed PZTNB-1 and YIG powders allows to significantly reduce content of fluorite-like foreign phase based on zirconium oxide, which forms due to the interfacial interaction during heat treatment and becomes stabilized by yttrium oxide. Connectivity has considerable effect on the value of ME coefficient of composite ceramics. With the same x value, $\Delta E/\Delta H$ characteristic decreases when changing from 0–3-type structured composites (PZT grains embedded in ferrite matrix) to 3-3-(interpenetrating network of two phases) and especially 3-0-type samples (YIG grains embedded in PZT matrix); in the last case this can be attributed to the substrate clamping effect when ferrite grains are clamped with piezoelectric matrix. $\Delta E/\Delta H$ value of 0–3 composites with $x=40$–60 wt% was found to be ~1.6 mV/(cm Oe).

1. Introduction

Magnetoelectric (ME) composite materials in which yttrium iron garnet (YIG, $Y_3Fe_5O_{12}$) is used as a magnetostrictive component and similar materials are representatives of two-phase multiferroic systems. They exhibit novel properties (product properties, [1–4]) which are not typical for individual phases of the material. These properties is a result of ensemble interactions between phases caused by the mechanical strain transfer from magnetostrictive phase to the adjoining piezoelectric phase (or vice versa) under the influence of an electric field.

Soft magnetic spinel ferrites are the most commonly used magnetostrictive materials for particulate ME composites [5–16]. On the other hand, extremely narrow ferromagnetic resonance absorption (FMR) line [17], which makes YIG unique among magnetoactive materials, allows to expect achievement of composites with high values of resonance ME effect that means the FMR line shift under the influence of an electric field. Other advantages of YIG include its record high specific electrical resistivity (up to 10^{13} Ω cm) that reduces eddy current loss during exploitation and provides high electrical resistivity of the resulting YIG ME composites, which is an indispensable condition for effective poling of piezoelectric phase.

The unique combination of magnetic, electric and ME properties opens new opportunities for exploitation of multiferroic heterostructures in such UHF devices as filters, attenuators, resonators, phase shifters, and others. Their advantages include high productivity, compactness, and compatibility with integrated circuit technology. The application of ME composites in resonance UHF systems allows to configure the device settings by electrical action.

Manufacturing of particulate piezoelectric/magnetostrictive ferrite ME composites is known to require high-temperature calcination of the mixture of powdered piezoelectric and magnetostrictive materials. Under these conditions, chemical interaction between components in the doping level is highly probable, and, in some instances, it may cause the formation of the new phases, especially in the case of YIG-containing ME ceramics. Thus, our study [18] shows that formation of foreign phase with pyrochlore structure in $PbTiO_3$/YIG system is a result of high-temperature calcination. At the same time, there is no chemical interaction in $BaTiO_3$/YIG system up to 1200 °C. However, elevation of the temperature up to 1300 °C causes decomposition of $Y_3Fe_5O_{12}$ into yttrium orthoferrite ($YFeO_3$). Well sintered high quality ceramic with no foreign phases was obtained using $Ba_{1-x}Pb_xTiO_3$/YIG composition with $x \le 0.2$ [18]. $Ba_{0.9}Pb_{0.1}TiO_3$/YIG composites exhibit the highest piezoelectric properties induced by effective poling owing to the relatively low ferroelectric rigidity of piezoelectric material and high specific electrical resistivity.

The data obtained in [18] correlates with the results presented by Yang et al. in work [19], which studies $BaTiO_3/BiY_2Fe_5O_{12}$ particulate composites obtained through standard solid-state method

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at 1100 °C. Similarly to $BaTiO_3$/YIG system, in $BaTiO_3/BiY_2Fe_5O_{12}$ specimens phases of barium titanate and ferrite coexist separately, no impurities were observed. Composites exhibit low leakage current loss, their ME behavior depends strongly on the concentration of $BiY_2Fe_5O_{12}$ and on the strength of static magnetic field. Maximal ME coefficient value reaches approximately 0.21 mV/(cm Oe). Far greater ME coupling efficiency can be achieved in laminate $BaTiO_3/BiY_2Fe_5O_{12}$ heterostructures [20]: value of their $\Delta E/\Delta H$ coefficient was found to be 14.36 mV/(cm Oe).

YIG-containing systems with lead zirconate titanate (PZT) in piezoelectric component are also the specimens of ME ceramics in which formation of fluorite-like foreign phase of zirconium oxide stabilized by yttrium oxide occurs when the material is calcined at a high temperature. According to [21], X-ray powder diffraction (XRD) study of YIG/PZT-36, YIG/PZT_TBS-2, and YIG/PZTNB-1 composite ceramics obtained at 1200 °C clearly defined pronounced reflections of the foreign phase. Clear evidence of the new phase was already encountered at 1050 °C. In the same time, since PZT materials are known to outperform barium titanate in piezoelectric output, the use of PZT in a piezoelectric component of ME ceramics based on YIG could increase ME coupling efficiency, provided that unwanted interfacial reactions are prevented.

The use of the finely dispersed powders of components that significantly decreases sintering temperature may be one of the means to reduce unwanted interfacial interaction. Sol–gel methods can be applied to prepare these powders. As another advantage, the use of the powdered components with submicron particle size gives possibility to obtain composite ceramics with different connectivity patterns (0–3 (piezoelectric grains embedded in ferrite matrix), 3–0 (vice versa), or 3–3 (interpenetrating network of two phases)) not only by altering the composition but also with the same composition by varying the fabrication conditions.

In this study we report the results of the investigation of $(100-x)$ wt% PZTNB-1 + x wt% YIG manufactured through various procedures including sol–gel-process. PZTNB-1 was selected because of its high efficiency (electromechanical coupling coefficient $k_p=0.6$, permittivity $\varepsilon/\varepsilon_0=1400$, piezoelectric coefficient $d_{33}=330$ pC/N, piezoelectric voltage coefficient $g_{33}=27$ mV m/N) and the ease of fabrication through sol–gel process [8].

2. Material and methods

$(100-x)$ wt% PZTNB-1 + x wt% YIG composites were prepared in three ways: (1) From PZTNB-1 and YIG powders synthesized through solid-state method and previously calcined at 1200 and 1250 °C, respectively; sintering mode: 1200 °C, within 2 h; (2) From PZTNB-1 powder synthesized through sol–gel process at 700 °C in accordance with technique described in [8] and from YIG synthesized through solid-state method at 1250 °C; sintering mode: 980 °C, within 2 h; (3) From PZTNB-1 synthesized through solid-state method and previously calcined at 1200 °C and from YIG synthesized through sol–gel method at 800 °C in accordance with the technique described in [22,23]; sintering mode: 1030 °C, within 2 h. It was expected that the use of components synthesized through sol–gel methods would allow to obtain high quality ME ceramics at a lower temperatures for the purpose to prevent interfacial interaction. Furthermore, procedures 1–3 with the average values of x would provide different connectivity: 3–3 by procedure 1, 3–0 and 0–3 by procedures 2 and 3, respectively.

The samples of $(100-x)$ wt% PZTNB-1 + x wt% YIG ME ceramics obtained by sintering were cut into disks, abraded and coated with silver paste which was burned in at 650 °C. Pulse polarization was applied to the samples within 3 min in chloroform medium in a field of 1.5–3 kV/mm within 3 min.

To ensure that the synthesis of PZTNB-1 and YIG has gone to completion and to define the phase composition, samples were studied by X-Ray powder diffraction (XRD) analysis using ARL-X'TRA diffractometer with CuKα radiation.

Morphology of fractured surface was studied by scanning electron microscopy (scanning electron microscope JEOL JSM 6390LA).

Dielectric and piezoelectric properties were studied by 《Cenzurka M》 computerized measurement system. Piezoelectric coefficient d_{33} was measured using quasi-static method by d_{33} meter. Electrical resistivity was measured by E6-13A teraohmmeter with direct current. ME properties of ceramics were studied at 1 kHz frequency under the simultaneous influence of the static (1–1.2 kOe) and varying (19.2 Oe) magnetic field by specific device (the scheme is given in [24]).

3. Results and discussion

Fig. 1a–d shows XRD patterns of $(100-x)$ wt% PZTNB-1 + x wt% YIG composites prepared through procedure 3. Patterns of the samples with equal weight ratio of components ($x=50$ wt%) prepared through procedures 1–3 are depicted in Fig. 1e–g. XRD patterns of pure PZTNB-1 and YIG materials (Fig. 1h and i, respectively) are also shown. Composites prepared through procedure 3 were used as an example to illustrate variations of aspect ratio of reflections intensities with changes in weight ratio. All samples, regardless of their production way, contain small amounts of fluorite-like foreign phase of zirconium oxide stabilized by yttrium oxide, this is characteristic feature of YIG-containing composites based on PZT [21]. However, amount of foreign phase is negligible in samples prepared through procedures 2–3 owing to the decrease in sintering temperatures by the use of finely dispersed PZTNB-1 and YIG powders. It should be noted that reflections of PZT phase are slightly shifted relative to the peaks of pure piezoelectric material, and in comparison with PZTNB-1 their

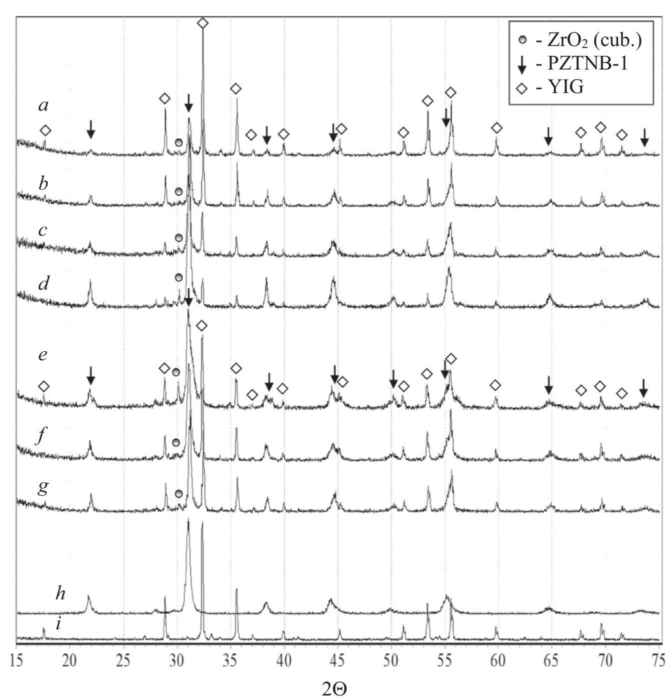

Fig. 1. XRD patterns of $(100-x)$ wt% PZTNB-1 + x wt% YIG system with x = (a) 20, (b) 40, (c) 60, (d) 80, (e–g) 50, (h) 0, (i) 100 prepared through procedures (e) 1, (f) 2, and (a–d, g) 3.