Magnetoelectric biodegradable uniform composite microactuators for biomedical applications

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Investigation of magnetostriction of the nanorods

The magnetostriction measurements were carried out using well-known strain gauge method, which often used for alloys and ceramic samples. For magnetostriction measurements of ferrite nanoparticles the method was adopted using technique described elsewhere.[1] For this procedure, the powder of Fe_3O_4 nanorods (NRs) with mass about 150 mg was pressed into disc shape pellets with a diameter of 10 mm under uniaxial pressure ~1 GPa, using cold powder pressing method. In order to preserve the initial properties of the powder as much as possible, no binder or high-temperature sintering was used. For better observation of magnetic anisotropy, the powder pressing process was combined with a magnetic field ~0.2 T applied to disc shape sample in plane direction (fig. SI1).

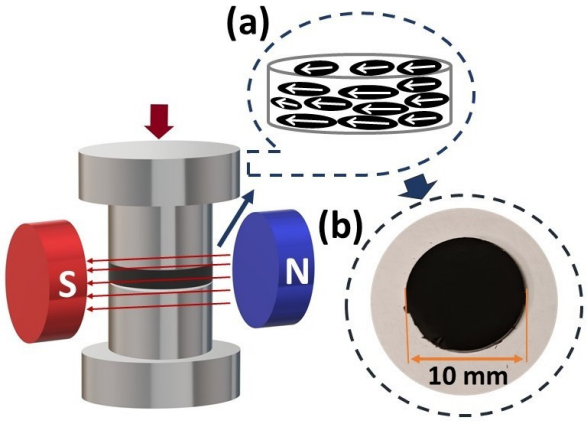

Figure SI1. Scheme of sample preparation.

For magnetostriction measurements the bi-directional strain gauge (Model SK-06-030TY-350, Micro-Measurements,Vishay Precision Group, Inc., Germany, 350 Ω) was used as a sensor, which allows to measure the magnetostriction in two directions: in longitudinal ($\lambda_{\parallel}$) and transverse ($\lambda_{\perp}$) to the direction of the applied magnetic field. Gauge was mounted on side of the sample is shape of disc using special adhesive based on polyvinyl butyral (BF-2, Solins, Russia). For gauge connection, the same temperature protocol for drying of bonded gauge was used (80 °C for 2 hours, followed by slow cooling for 12 h). Magnetostriction was measured at room temperature (~295 K), when the magnetic field was applied in plane of the sample. The system of NdFeB

permanent magnets (N35) constructed to Hallbach type structure with maximum of magnetic field ~0.5 T was used as magnetic field source.

As is known, the magnetostriction refers to magnetically elastic phenomena and reflects the relationship between magnetic and elastic properties in ferromagnets, due to changes in the magnetic moment and exchange interactions, which are interrelated with the magnetocrystalline anisotropy. The magnetostriction of iron oxide Fe_3O_4 is anisotropic and depends on the magnetic domain structure of the sample.[2] As a result, the magnitude of the magnetostriction of Fe_3O_4 with different types of crystal structure (single crystal, polycrystalline ceramics) can strongly depend on the technological conditions of synthesis. In particularly, a dependence of the magnetostriction on the size of Fe_3O_4 nanoparticles was observed: an increase in the size of nanoparticles leads to an increase in the magnetostriction coefficient.[1] Our measurements demonstrate that the Fe_3O_4 NRs have longitudinal $\lambda_{\parallel}$=−16 ppm and transverse $\lambda_{\perp}$=4 ppm magnetostriction coefficient at applied magnetic field 0.5 T. According to the theory[3] in the region of magnetic saturation relationship $\lambda_{\parallel} = -2\,\lambda_{\perp}$ is fulfilled for polycrystalline samples. However, in our case, this relation is not fulfilled, which indicates a more pronounced anisotropic type behavior of Fe_3O_4 NRs. The observed value of $\lambda_{\parallel}$ as was expected is larger, than for Fe_3O_4 nanoparticles: from −6.5 to −14.6 ppm[1] and less than for Fe_3O_4 single crystals: about −20 ppm[4], which confirms the role of magnetic anisotropy. The volume magnetostriction of Fe_3O_4 nanoparticles, defined as $\lambda\parallel + 2\,\lambda\perp$, is −8 ppm, which significantly exceeds the similar values for Fe_3O_4 nanoparticles: −3.4 ppm.[1] Thus, as can be concluded from magnetostriction measurements, the nanoparticles in the shape of NRs can be proposed as one of the ways to enhance the magnetoelectric effect in multiferroic nanocomposites.

The measurements of the magnet magnetic field strength

We have measured the magnetic field strength of the magnet using a Hall sensor both at room temperature and at 200°C. As shown in figure SI2, the field strength at the sample location is 50±10 mT, and it remains stable even at the elevated crystallization temperatures for poly(L-lactic acid) (PLLA) (170–200°C). At 300°C, the field strength at this location decreases by approximately 15% and does not fully recover upon cooling.

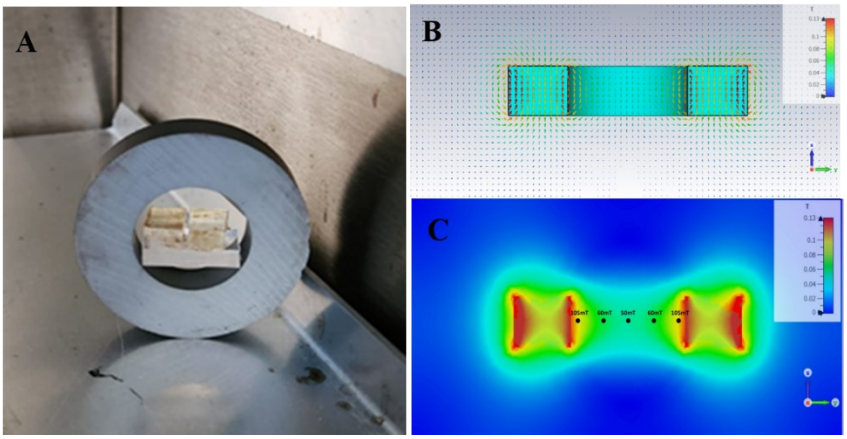

Figure SI2. Characterization of the magnetic field strength. (A) The photo of sample location relative to the magnet. Direction (B) and (C) amplitude of the magnetic field vector. The color bar indicates the field strength in mT. Data was collected at 25°C and was verified to be stable up to 200°C.

AFM measurements

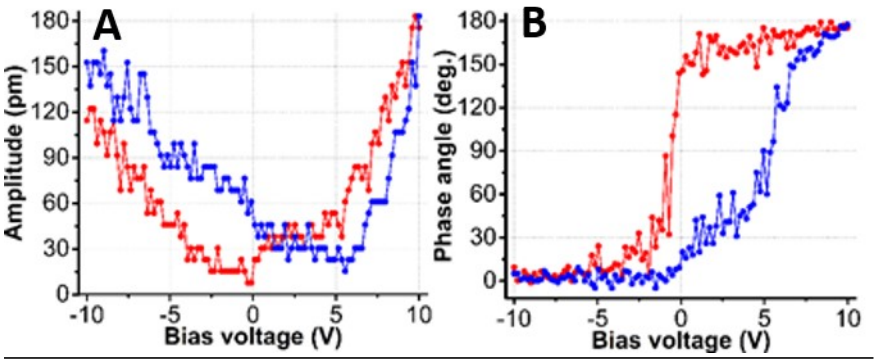

Figure SI3. Graphs of the dependence of the change in the amplitude (A) and phase (B) of the local piezoelectric response signal on the applied voltage.

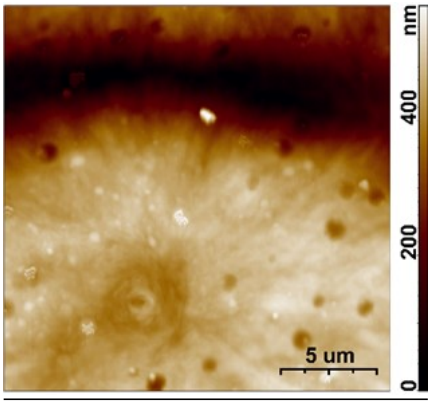

Figure SI4. The AFM image of the studied surface area of crystallized PLLA/NRs composite.

The vertical magnetic field (MF) applied during PFM-signal removal induces an "out-of-plane" effect (perpendicular to the substrate plane) on the NRs (Fig. SI5).

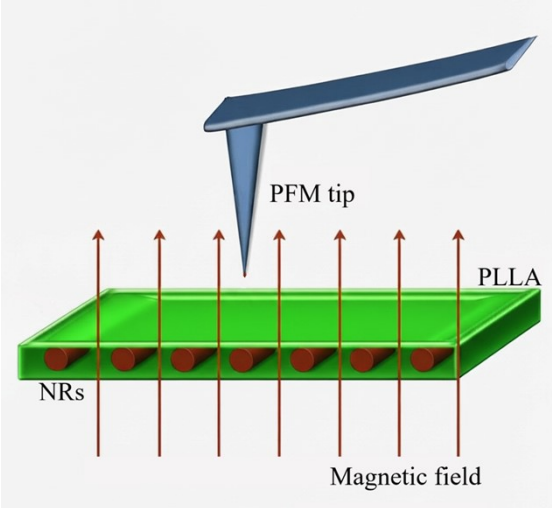

Fig.SI5 Schematic representation of PFM measurements in the presence of a vertical MF.

Consequently, the mechanical stress experienced by the NRs is transmitted primarily along this axis to the PLLA matrix, inducing a mechanical deformation directed toward the probe. This was demonstrated in an experiment where a 1000 G horizontal magnetic field (MF) was applied during the removal of the piezoresponse force microscopy (PFM) signal, as illustrated in the figures below. To obtain statistically significant data, we measured 10 points on the PLLA/NRs surface. The average d_{33} value was 14.84 ± 2.93 pm/V in the absence of a magnetic field (MF), and increased to 18.13 ± 3.61 pm/V upon application of a 1000 G MF (fig. SI6).

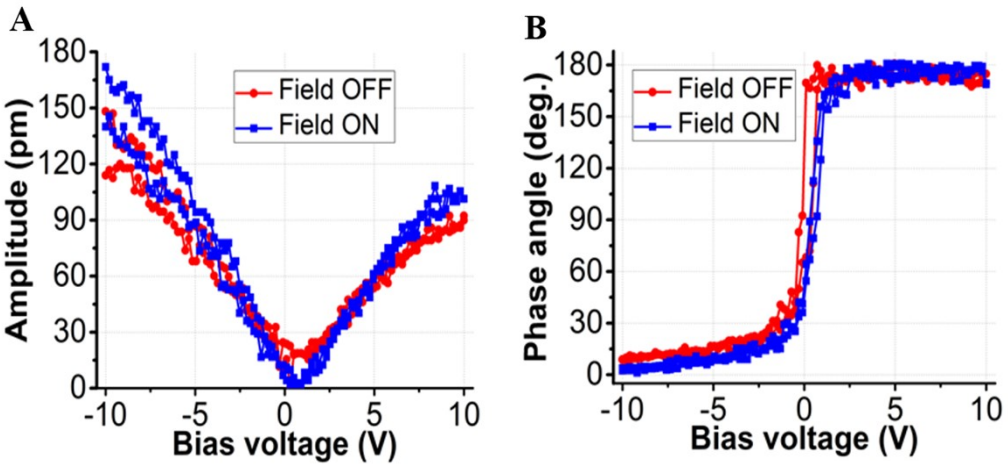

Figure SI6. Graphs of the dependence of the amplitude (A) and phase (B) changes in the PLLA/NRs matrix when exposed to an external horizontal magnetic field of 1000G

The experimental results demonstrate that the application of a horizontal magnetic field does not significantly alter the measured d_{33} coefficient. This observation can be attributed to the direction of the induced mechanical deformation, which is oriented primarily toward the PLLA matrix and consequently has a negligible impact on the PFM probe during signal acquisition.

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