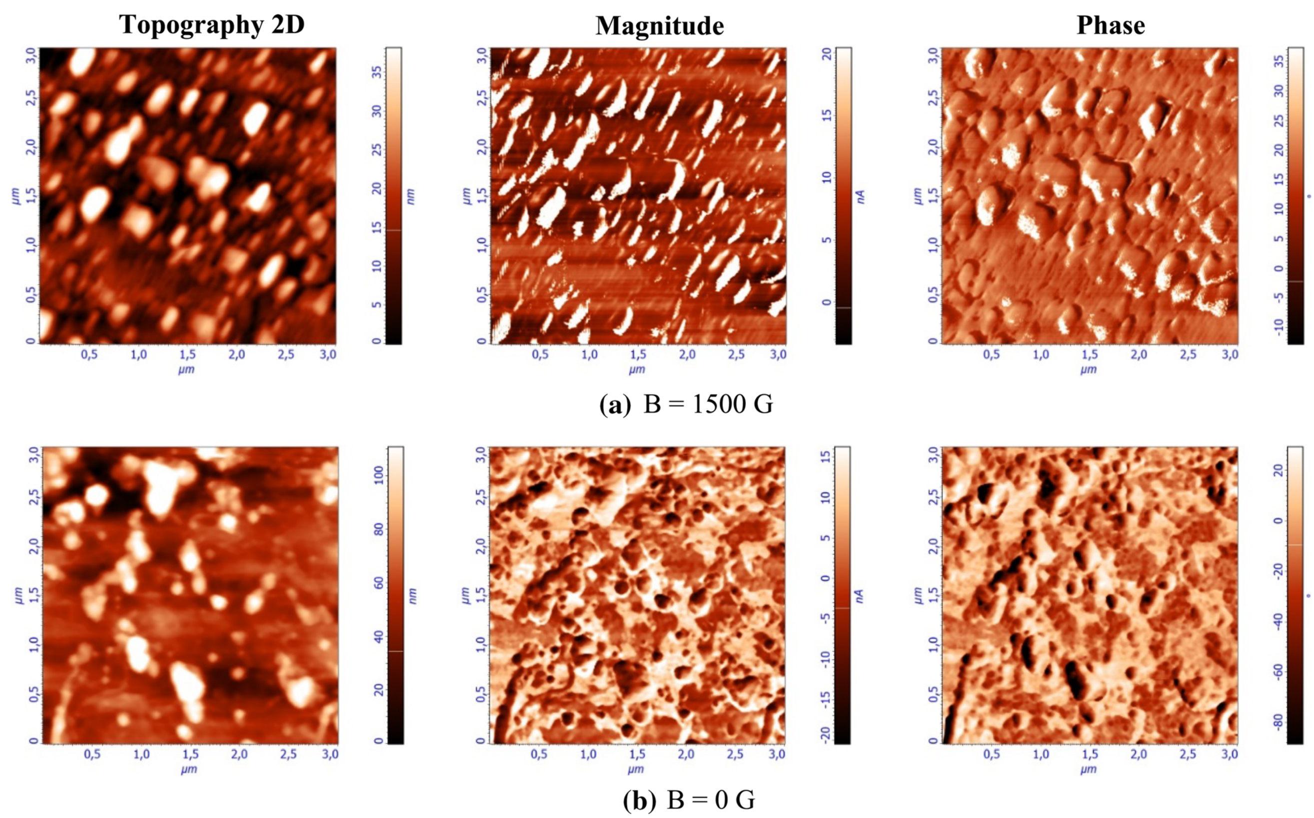

Figure 12 Comparative PFM images showing the surface topography 2D, the magnitude and the phase of the piezoresponse for x = 4 vol% BFT composite: **a** under a magnetic field application of B = 1500 G and **b** after magnetic field application.

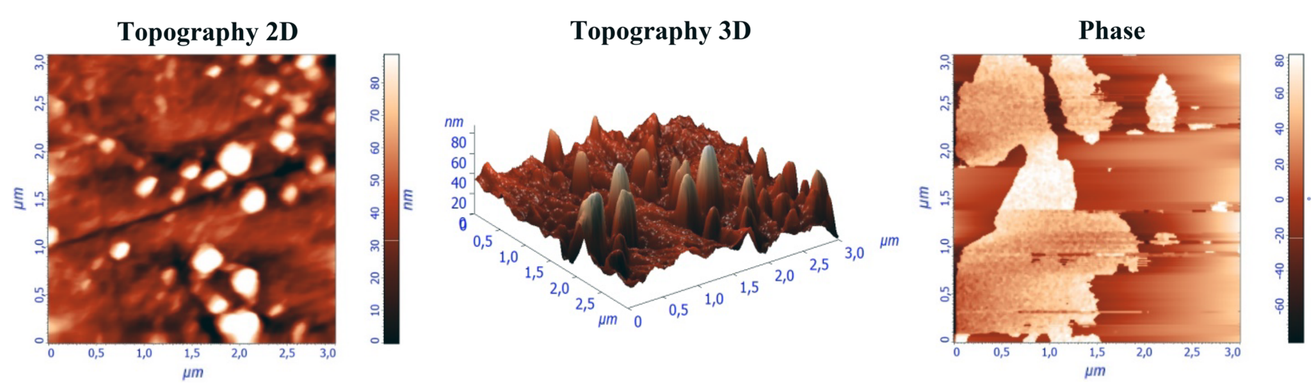

Figure 13 MFM images showing the surface topography and the phase of the magnetic response for x = 4 vol% BFT composite.

enhancement or the cancellation of the local magnetic field strength due to interaction between magnetic particles with different orientations.

Local PFM and MFM investigations have shown a combined ferro/piezoelectric character and magnetic order, with magnetoelectric coupling demonstrated by the reorientation of filler particles and modifications of local piezoresponse when applying a static magnetic field.

In conclusion, the results from electrical, magnetic, ferroelectric and PFM–MFM experiments are well correlated and demonstrate that $(1-x)$PVDF–$(x)Ba_{12}Fe_{28}Ti_{15}O_{84}$ composites are magnetodielectrics with both magnetic and ferro/piezoelectric properties, making the system suitable for multifunctional materials applications.

Conclusions

In this paper, $(1-x)$PVDF–$(x)Ba_{12}Fe_{28}Ti_{15}O_{84}$ flexible composite thick films with $x < 9$ vol% were prepared and investigated at various length scales. The X-ray diffraction and FTIR patterns confirm the formation of PVDF α-crystalline phase with small amount of polar phases, e.g., γ-phase, which is the polar phase with the lowest dipole moment. The SEM and AFM images show that the filler particles are homogeneously distributed inside the PVDF polymer matrix. A predominant soft magnetic character derived from the filler properties is observed in the $(1-x)$PVDF–$(x)Ba_{12}Fe_{28}Ti_{15}O_{84}$ composites, with a maximum value of M_s = 2.6 emu/g for the highest concentration of 9 vol% and a very low coercivity of $\sim$ 60 Oe. The room-temperature permittivity of PVDF-based composites presents a modest increase with BFT addition and remains below 10 for all the filler volumes. All the composites present similar activation energies of 0.68 eV that might be associated with the crystalline relaxation in α-phase of PVDF matrix. The P–E hysteresis loops are non-saturated and show that the BFT particles contribute to a small increase in the effective polarization in composites. The PFM and MFM experiments showed that $(1-x)$PVDF–$(x)Ba_{12}Fe_{28}Ti_{15}O_{84}$ composites present both magnetic and ferroelectric properties, which are also coupled via strain–stress mechanism. In conclusion, $(1-x)$PVDF–$(x)Ba_{12}Fe_{28}Ti_{15}O_{84}$ composites with small filler additions $x \leq 9\%$ are magnetodielectrics with reasonable dielectric, ferro/piezoelectric and soft magnetic character, while maintaining flexibility.

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