## Dielectric and magnetic properties in Co- and Ni- containing ferrite/poly(vilidene fluoride) multiferroic nanocomposites

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## **Abstract**

Particulate composites films of poly(vinylidene fluoride) and  $CoFe_2O_4$  and  $NiFe_2O_4$  ferrites (supplied by Nanoamor, grain sizes between 35-55 nm and 20-30 nm for the Co- and Ni-containing ferrite, respectively) were prepared from solvent casting and melt processing. The weight percentage of ferrite nanoparticles varied from 0.001 to 50 in the case of Co-ferrite and 5 to 50 in the case of Ni-ferrite.. X-ray diffraction studies demonstrate that ferrite nanoparticles nucleate the piezoelectric  $\beta$ -phase of the polymer, but the different ferrites nucleates the whole polymer phase at different concentrations [1] (see Fig. 1).

The macroscopic dielectric response of the composites demonstrates a strong dependence on the weight fraction of ferrite nanoparticles. In all cases an increase of the  $\epsilon$ ' for the composites with respect to the pure polymer sample has been observed. The inclusion of ferrite nanoparticles in the PVDF actually nucleates the  $\beta$ -phase of the material as the characteristics parameters of the  $\beta$ -relaxation of the polymer are maintained with respect to the values obtained for  $\beta$ -phase obtained by stretching from the  $\alpha$ -phase material, as analysis through the Vogel-Fulcher-Tamann (VTF) formalism demonstrates (see Fig. 2).

Low field ZFC-FC magnetization measurements reveal a remarkable similarity for both pure ferrites. However, room temperature measured hysteresis loops for the pure ferrites and also composites with different percentages of ferrite nanoparticles inside, clearly show superparamagnetic behavior for NiFe<sub>2</sub>O<sub>4</sub>/PVDF composites, while the CoFe<sub>2</sub>O<sub>4</sub>/PVDF samples develop an hysteresis cycle with coercivity of 0.3 T (see Fig. 3). In all cases the fit of magnetization data at high fields allow us to deduce values of the anisotropy constant  $K_{eff}$ , being this of 1.58 x 10<sup>5</sup> erg/cm<sup>3</sup> and 0.12 x 10<sup>5</sup> erg/cm<sup>3</sup> for the CoFe<sub>2</sub>O<sub>4</sub>/PVDF and NiFe<sub>2</sub>O<sub>4</sub>/PVDF composites, respectively. Finally, The shape of the measured M(H) loops with different directions (in-plane and perpendicular to the plane of the composite) of the applied magnetic field also demonstrates that magnetic particles are randomly oriented within the polymer matrix. This fact is fully supported by the observed linearity in M<sub>s</sub> value vs. % ferrite content, hinting for an homogeneous distribution of the ferrite nanoparticles whithin the composites.

## References

[1] P Martins, C M Costa, G. Botelho, S Lanceros-Mendez, J.M. Barandiarán and J. Gutiérrez, Materials Chemistry and Physics, **131** (2012) 698-705, and references therein.

## **Figures**

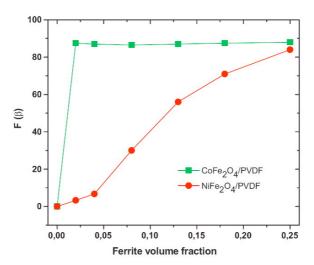


Figure 1.  $\beta$ -Phase content of the PVDF nanocomposites as a function of CoFe $_2$ O $_4$  and NiFe $_2$ O $_4$  ferrite content.

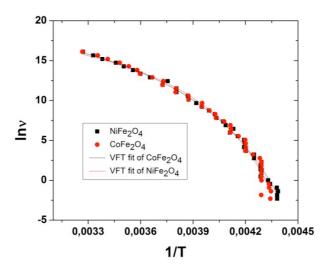


Figure 2. VTFH fittings to the  $\beta$ -relaxation for CoFe<sub>2</sub>O<sub>4</sub>/PVDF and NiFe<sub>2</sub>O<sub>4</sub>/PVDF composites with 20% of ferrite content.

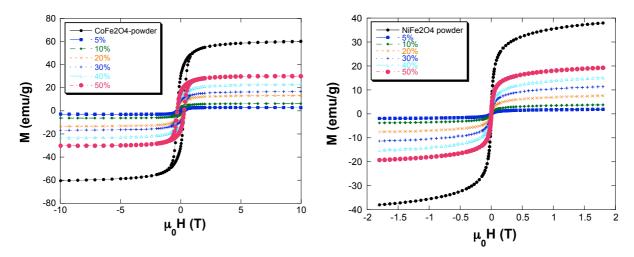


Figure 3. Room temperature hysteresis loops measured for (*left*) CoFe<sub>2</sub>O<sub>4</sub>/PVDF and (*right*) NiFe<sub>2</sub>O<sub>4</sub>/PVDF nanocomposites with different ferrite concentrations. Hysteresis loops for pure ferrites are also shown.