Robust piezoelectric coefficient recovery by nano-inclusions dispersion in un-poled PVDF-$Ni_{0.5}Zn_{0.5}Fe_2O_4$ ultra-thin films

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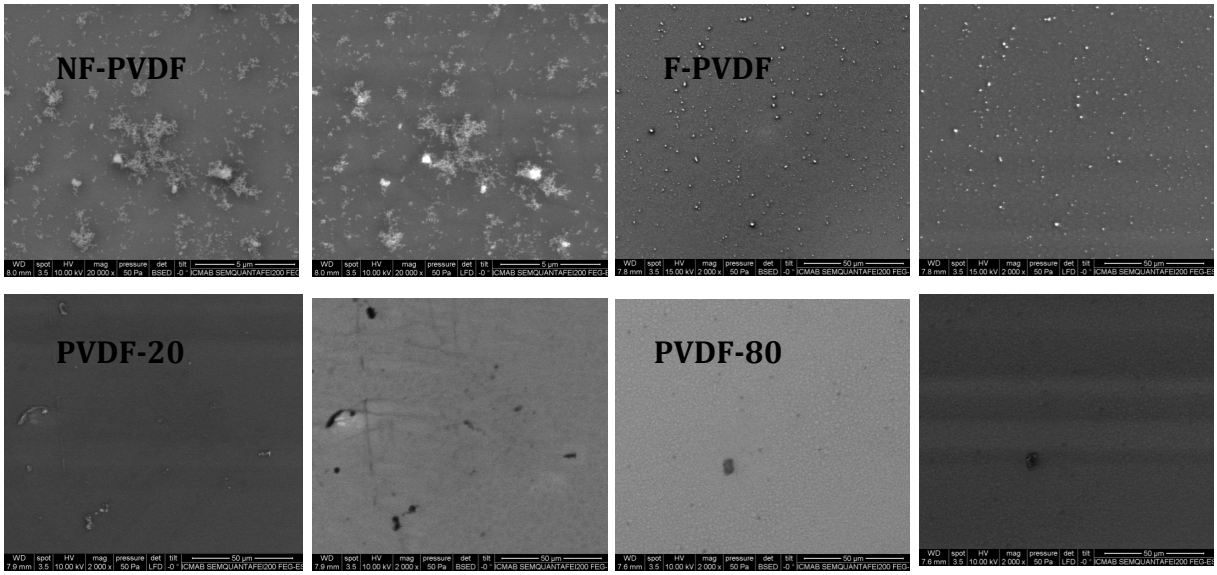
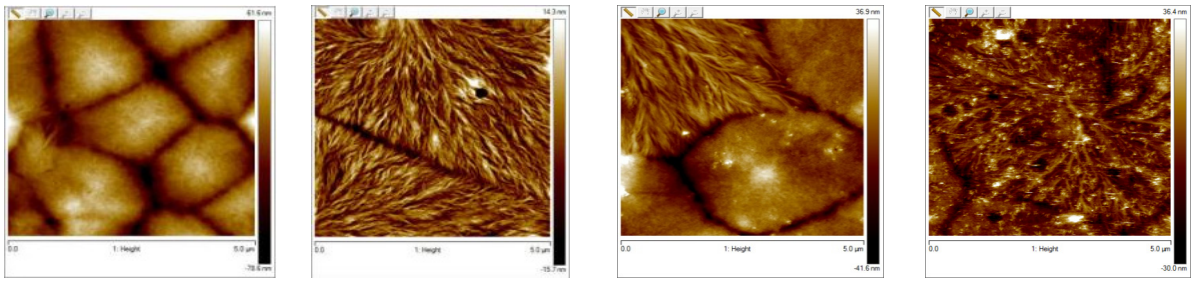
Featured Application: shapeable piezo-magnetic sensors.



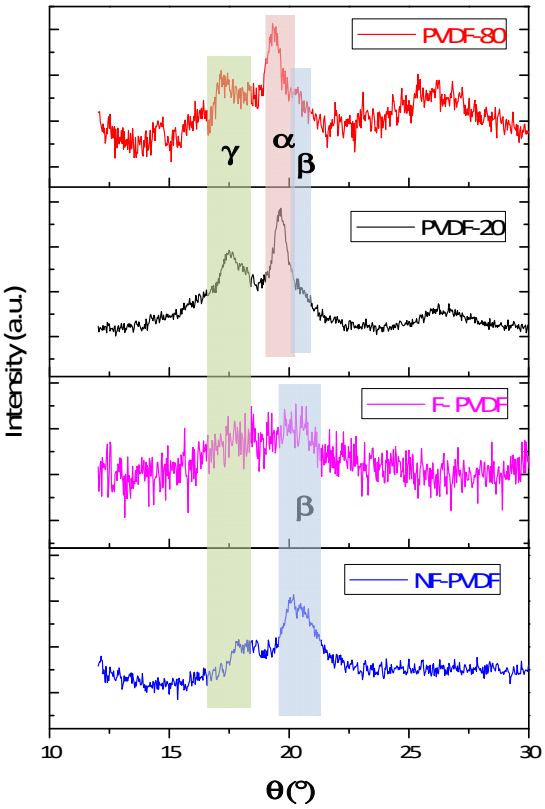
Supplementary Materials Figure S1: From left to the right and top to bottom: The SEM images of non-functionalized PVDF hybrid film (NF-PVDF), functionalized PVDF hybrid film (F-PVDF), PVDF neat film elaborated at 20°C (PVDF-20) and PVDF neat film elaborated at 80°C (PVDF-80). The Back Scattered Electron Detector-BSED (labled images) and Large Field Detector-LFD (on the right side of each labled images) are used to confirm the difference phases in samples.



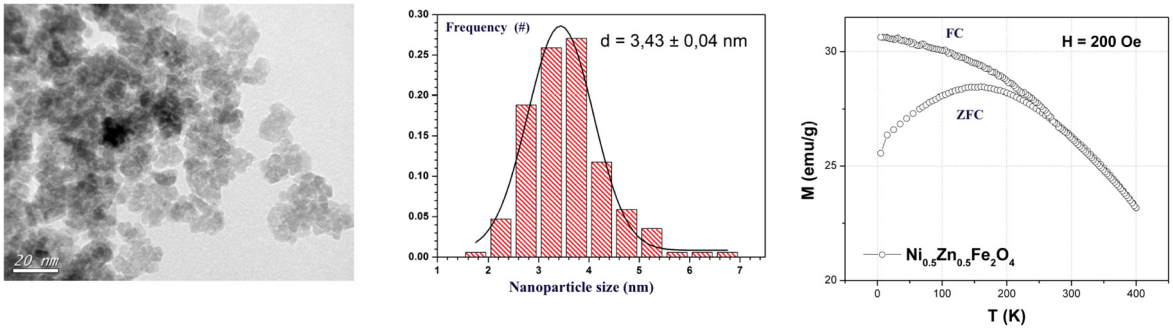
Supplementary Materials Figure S2: From the left to right: topography images of PVDF-20, PVDF-80, NF-PVDF and F-PVDF samples. The topography of our thin film was measured by a standard AFM imaging analysis. (Technical details: CSC38/Cr-Au including 3 cantilevers, using contact mode, resonance frequency 10 kHz, 14 kHz, 20 kHz). Spherulitic micrometric structures as well as the sub-micron fiber ones are clearly visible and in agreement with the previous observations. Images are 5µmx5µm sized.

Sample	PVDF-20	PVDF-80	NF-PVDF	F-PVDF
Substrate	Si	Si	Si	Si
Substrate Temperature	20°C	80°C	20°C	20°C
DMF/Acetone concentration	1:1	1:3	1:1	1:2
Nano-Particles (wt% and Functionalization)			0.5%NF-NPs	0.5%F-NPs
Thickness (nm)	99±2	83.3±1.5	75.7±3.7	80±4
Mean Roughness (nm)	14.5±0.1	3.5±0.1	5.5±0.1	6.3±0.1

Supplementary Materials Table S1: Substrate temperatures and solution concentration used in the elaboration process are reported from [42]. The overall topography roughness was statistically determined and reported here together with the thickness values.

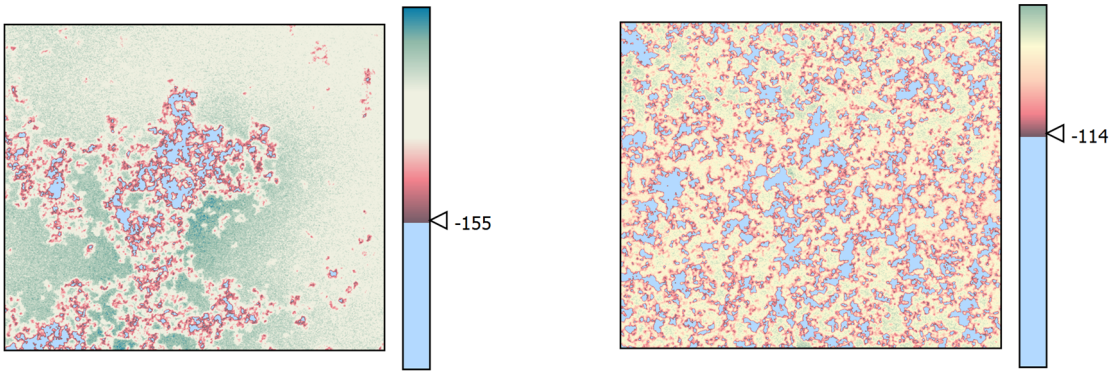


Supplementary Materials Figure S3: X-Ray diffraction patterns acquired on the studied four samples. The main Bragg peaks of the PVDF crystalline phases are evidenced. Diffraction lines associated with nanoparticles crystalline peaks concerns higher angles than the one reported here and have already been analysed and reported in reference 42 (figure 6 of the article) for both F-PVDF and NF-PVDF thin films. They are mono-crystalline and present the good spinel symmetry.



(a) (b) (c)

Supplementary Materials Figure S4 : (a) TEM images of the nanoparticles showing the spherical morphology; (b) statistical analysis of the nanoparticles size showing a diameter of 3,5 nm; (c) nanoparticles magnetization as function of the temperature measured after a Zero Field Cooled (ZFC) procedure and during the colling down under the application of a magnetic field (Field Cooled) of 200 Oe. The magnetization put in evidence a blocking temperature of 250K typical of the superparamagnetic phenomenon well known in the case of nanometric ferromagnetic nanoparticles. This high value of blocking temperature is also a demonstration of the mono-crystalline quality of the spinel nanoparticles and their dispersion (see for example A. G. Kolhatkar, A. C. Jamison, D. Litvinov, R. C. Willson and T. Randall Lee, *"Tuning the Magnetic Properties of Nanoparticles"*, Int. J. Mol. Sci. 2013, 14, 15977-16009; doi:10.3390/ijms140815977)



Supplementary Materials Figure S5: *ImageJ* processing and analysis of the SEM images of (a) NF-PVDF film and (b) F-PVDF one. The analysis of the nanoparticles agglomarates have shown 21 similar agglomarates in the first case compared to 77 in the second one. The surface of the identified agglomarates in NF-PVDF film is 4 times bigger than the one of the F-PVDF sample. Colour bar indicates the blue area (with red edges) considered for counting and sizing the nanoparticles agglomarates.