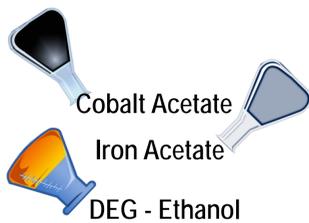


## Abstract

The magnetoelectric (ME) composites with piezoelectric and magnetostrictive material are of interest in the smart manufacturing and mechatronics fields as actuators/transducers, sensors, antennas, filters, non-volatile memories, etc. Spinel cobalt ferrite (CFO), a highly magnetostrictive material and niobium-doped lead zirconate titanate (PZTN), a piezoelectric perovskite phase are chosen as constituent phases for ME composite. The production process is designed in order to avoid the chemical reaction between the piezoelectric and magnetostrictive materials, to prevent the formation of percolation chains of the magnetostrictive phase, and to maximize the mechanical coupling at the interface between the two phases. In this view, the electrophoretic deposition (EPD) is a low cost and flexible technique to shape nanoparticles into multilayered heterostructures. The combination of different materials by EPD, showing promising ME coupling, can be regarded as a useful, preliminary approach in the search of novel ME materials for many applications, potentially with great industrial and technological benefits. In this work, composite bilayer CFO/PZTN thick films were deposited on platinum coated alumina by EPD from ethanol-based colloidal suspensions. Good adhesion and compaction of the green film were achieved by optimization of deposition voltage and time, and high density of the film and minimized interphase reactions occurred after sintering. The chemical activity between the two layers was controlled through the batches composition and it could lead to the synthesis of complex engineered structures. The deposited volume, the mixing of dielectric and magnetic phases and the density and ordering of the films have been verified by electron scanning microscopy after heat treatment. The ferroelectric, piezoelectric and magnetic properties were tested on the sintered films.

## Suspensions

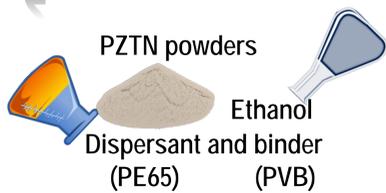
### 1 - Cobalt Ferrite [1-3]



Stoichiometry (spinel) :	$\text{CoFe}_2\text{O}_4$
Particles Density:	5.27 g cm <sup>-3</sup>
Suspension Density:	0.83 g cm <sup>-3</sup>
Solid Loading, wt%:	0.34
Particle Size (DLS):	10.1 nm
Viscosity:	56.7 mPa s
ζ-potential :	47.5 mV
Electrical Conductivity:	15 μS cm <sup>-1</sup>

- Solubilisation at 110°C for 1 h
- Heating to 180°C (2°C/min)
- 3h at 180°C • Air cooling to RT • Dilution in ethanol

### 2 - Lead zirconate titanate [4, 5]



Nominal stoichiometry (perovskite) :	$\text{Pb}_{0.988}(\text{Zr}_{0.52}\text{Ti}_{0.48})_{0.976}\text{Nb}_{0.024}\text{O}_3$
Particles Density:	~ 8 g cm <sup>-3</sup>
Suspension Density:	0.91 g cm <sup>-3</sup>
Solid Loading, wt%:	15.0
Particle Size (DLS):	185 nm
Viscosity:	1.08 mPa s
ζ-potential :	15.6 mV
Electrical Conductivity:	2 μS cm <sup>-1</sup>

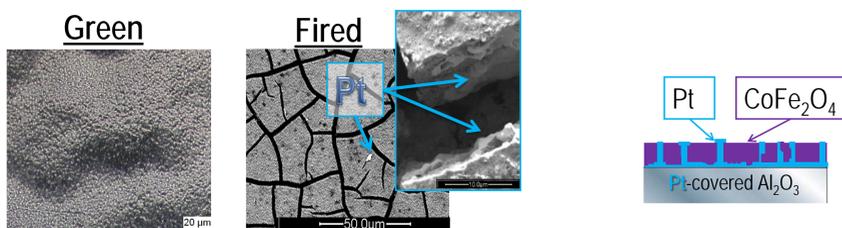
- Ball milling and stirring

## Electrophoretic Deposition [6]

EPD tests were performed in a plane-parallel cell geometry (1 cm electrodes spacing) and setting cathodic modality with constant DC potential up to 60 V vs. a 20 cm<sup>2</sup> SS secondary electrode.

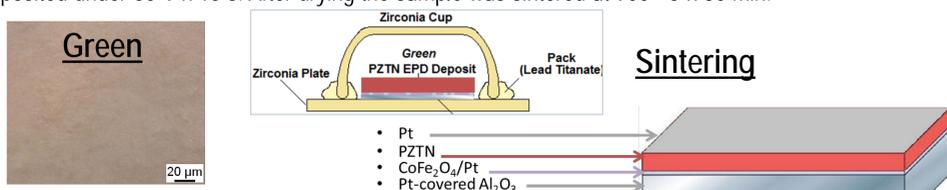
### 1 - First EPD-CFO film on Pt-coated alumina:

CFO suspension was deposited at 50 V x 100 s. After drying the sample was fired at 500 °C x 15 min.



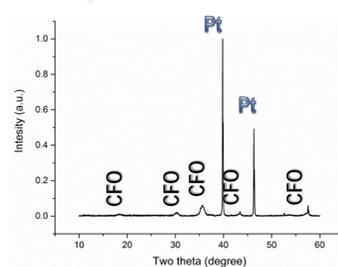
### 2 - Second EPD-PZTN film on first EPD-CFO layer:

PZTN suspension was deposited under 60 V x 15 s. After drying the sample was sintered at 900 °C x 30 min.

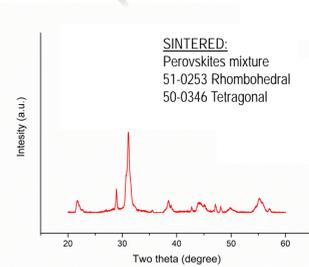


## XRD

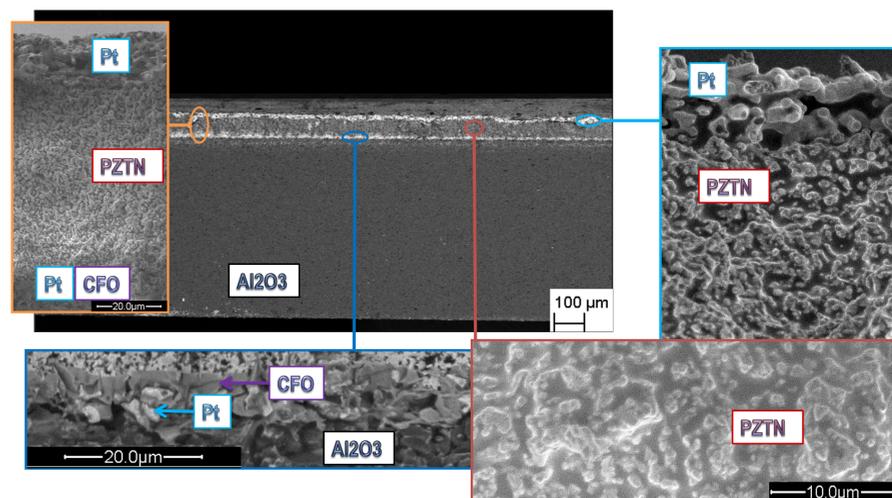
### 1 - Pt/CFO layer



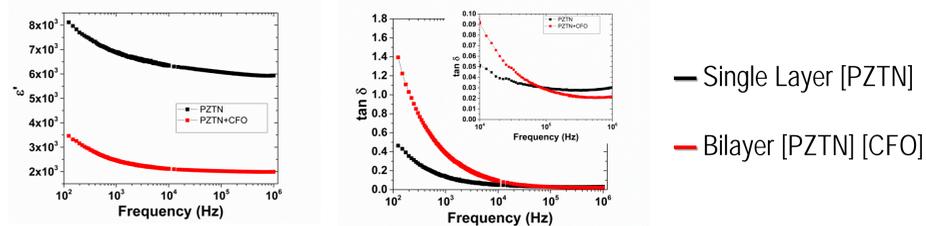
### 2 - PZTN layer



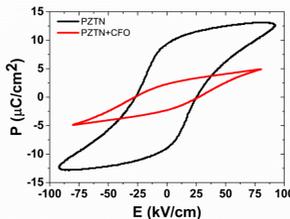
## SEM



## Dielectric characterization



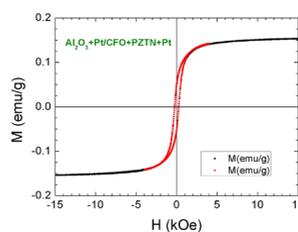
These values for the single layer PZTN are comparable to those obtained for Nb doped PZT bulk material. The CFO layer shows a detrimental effect on permittivity and losses, as expected from the mixing rule.



Hysteresis loop confirms the ferroelectric properties.

The observed distortion is probably associated with conductivity effects caused by the presence of the CFO magnetic phase.

## Magnetic characterization



Hysteresis loop confirms the ferromagnetic properties.

While the high coercive field is in agreement with the hard behaviour of the cobalt ferrite

## Conclusions

- Magnetolectric composite bilayer films based on spinel cobalt ferrite and perovskite Nb doped PZT was produced by electrophoretic deposition
- The microstructure analysis was performed by SEM/EDS and XRD
- The electrical and magnetic characterization confirm a good quality of the piezoelectric and magnetic phases, respectively
- Future work is going to develop the coupling between magnetic and electric ordering

## References

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