Theoretical optimization of magnetoelectric multilayer laminates

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Magnetoelectric (ME) materials are becoming increasingly relevant in the development of new technologies for biomedical applications, sensors and actuators, among others. Mathematical models and simulations allow to optimize features and acquire fundamental knowledge on material properties to achieve innovative developments and devices. In this way, this work is focused on the simulation of both polymer-based and ceramic-based ME laminates, in order to evaluate the influence of their structure, mechanical, electrical and magnetic properties on the ME response.

The effect of size and configuration has been evaluated in Vitrovac/poly(vinylidene fluoride)(PVDF) and Vitrovac/ lead zirconate titanate (PZT) laminated composites. It has been established that the elastic properties and amorphous constitution of PVDF are key parameters governing its ME response, increasing its influence with increasing number of layers in the composite. Although configurations of multi-layers where the laminate is sandwiched between magnetostrictive layers (M-M configurations) exhibited more stable ME response, the structural influence of PVDF in the configurations where the multi-layer is sandwiched between piezoelectric layers (P-P configurations) establishes peaks of ME performance. Good agreement is established when comparing trends reported experimentally in the literature for magnetostrictive to piezoelectric thickness ratio of multilayers, presenting a curve that rapidly increases their α_{unit} with increasing thickness ratio up to $n=0.3$, when saturation is reached. Further, an optimal configuration for PZT multilayers is found, with external magnetostrictive phases and thickness ratio above 0.2, leading to a ME response of 86.7 V/cm. Finally, it has been established that PVDF configurations with external magnetostrictive phases (M-M configurations) show more stable behaviour and trends over different number of layers, of about 11.5 V/cm, while P-P configurations present regions with peaks out of the expected trend with ME response (48 V/cm) closer to the one of ceramic multilayers .

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1. Introduction

Magnetoelectric (ME) and multiferroic materials are being increasingly studied from the fundamental and application point of view. The coupling between magnetic and electric properties allows to tackle novel scientific challenges and the development of multifunctional devices if the magnetic and electrical orders can be mutually controlled [1]. Further, the miniaturization of devices has guided the attention into a new generation of multifunctional materials, that allow a wide range of applications from broadband magnetic field sensors, actuators and memory devices, to magnetically controlled electro-optic or piezoelectric devices, among others [2, 3].

Single-phase materials with ferromagnetic and ferroelectric properties include XXX, but are limited in number [4] and present low ME performance and low Curie temperature[5]. Thus, efforts have been established to develop composites with higher ME coupling than single-phase materials[6]. Composites with magnetostrictive (ms) and piezoelectric (pzo) phases provide the best systems to improve ME response, and the knowledge of these materials from the fundamental point of view is the key tool to achieve optimized performances, being theoretical models and simulations a key element to optimize coupling and geometry [7, 8].

The ME coupling may arise by the effect of an electric (magnetic) field on the magnetization (polarization) of a material, or indirectly via strain over the magnetostrictive-electroactive domains of a ME composite [4]. The ME voltage coefficient (α_{ME}) is described as the change in electrical polarization, P , caused by the variation of the magnetic field, H , as[9]:

$$\alpha_{ME} = \frac{\delta P}{\delta H} \quad \text{Eq. 1}$$

The ME effect in composites is more than 2 orders higher than for single-phase materials[10] and can be found in magnetic metals/alloys and piezoelectric ceramic composites, laminated metals/alloys and piezoelectric polymers and particulate composites of ferrite and piezoelectric ceramics[5].

Ceramic ME composites exhibit much higher α_{ME} than polymer-based composites, but the latter have the advantage of a good strain coupling and durability, higher flexibility, without leakage currents, the possibility of being shaped into a variety of forms (sheets, moulded shapes), and the low temperature processing [5, 11]. Further, some of them are biocompatible [12]. Further, polymer-based composites avoid the limitations of the interface reactions on the ceramic composites that cause high dielectric losses and may affect the stability of the developed ME devices[5].

Polymer-based ME composites exist as laminates or nanocomposites (low dimensional-structured). Although low-dimensional structured composites present no interface degradation, laminate types of ME materials present a ME coefficient that can reach up to four orders of magnitude higher than the ones reported on nanocomposites [13]. ME composites with a laminated bi-layer or multi-layer configuration show also advantages such as avoiding the polarization loss in bulk composites[14]. The piezoelectric phase can be poled to enhance the ME coupling and it is also possible to change the poling and applied field directions to achieve maximum ME coupling[15]. Mathematical models and, particularly, Finite element method simulation models

represent a suitable tool to optimize the performance of such new materials [7, 8], by incorporating a wide spectrum of parameters, including physico-chemical characteristics of materials and geometries [7, 8]. This methodology is even more interesting when the ME performance of the materials are difficult to obtain by direct measurements and provide knowledge that can lead to the production of new and optimized materials for devices [16].

Previous works have been devoted to study the ME effect at the interface between the magnetostrictive and piezoelectric phases in low dimensional polymer based fibrillar [7] and spherical structures [8]. Similar mathematical models and simulations were used in order to study the ME effect at the interface between the magnetostrictive and piezoelectric phases.

Thus, this work reports on the multilayer structure optimization of ceramic-based and polymer-based composites, taking into consideration the effects of xxxxx, allowing not just the understating of the coupling between magnetostrictive and piezoelectric layers and their dependence on material characteristics, but also the design of optimized structures for device applications.

For the simulations, piezoelectric poly(vinylidene fluoride) (PVDF) was the selected polymer due to its high piezoelectric response, thermal, mechanical and chemical resistance [15, 17]. The piezoelectric ceramic material was PZT-5H, based on its piezoelectric response and complete characterization [18-24]. Vitrovac 4040® [25] ($\text{Fe}_{39}\text{Ni}_{39}\text{Mo}_4\text{Si}_6\text{B}_{12}$) was used as magnetostrictive component based on its high piezomagnetic coefficient at low magnetic fields [16].

2. Theoretical Models

2.1 Electromechanical finite element method model for ME laminates

As both magnetostrictive and piezoelectric behaviors are anisotropic and as the model incorporates specific mechanical coupling factors into the final ME response, finite element method (FEM) is a suitable approach for the theoretical study of laminated ME composites [16]. Assuming a stresses state on the magnetostrictive and piezoelectric phases, and linear range of magnetostriction, the electro-mechanical coupling was modeled following the main guidelines reported in references [7, 8, 16] and considering perfect bonding between interphases. Additionally, a 2D model was assumed considering the ME effect to be constant along the width of the structure. Based on the constitutive equations [26] the calculations were performed on each nodal point with specific boundary conditions and holding into two essential principles: energy conservation and continuity at the nodal interface. The electromechanical simulation consisted in applying a deformation on the lateral ends of the magnetostrictive layer taking into account the magnetostrictive response of the material [7, 8, 16, 25, 27] and evaluating the electric potential developed across the piezoelectric layer. The input parameter for the calculations was $S=\lambda(H)$, which is the magnetically induced magnetostrictive strain, the magnetostriction curve of the material. In all cases, it was selected the strain corresponding to the maximum deformation experienced by the magnetostrictive layer. Structurally, as the layers were perfectly bonded and

assuming linear range of magnetostriction, the deformation on the magnetostrictive layer produces a deformation on the other layers that will depend on their mechanical properties. Boundary conditions as lateral polarization of the piezoelectric material, grounds, and fixing the ME structure only to deform along the longitudinal direction were also set.

2.2 Mechanical model for the piezoelectric material

Constituent equations for the linear piezoelectric effect (in stress charge form) are given by the direct and indirect piezoelectric effects [28, 29], respectively:

$$\sigma_m = c_E \cdot S - e_s \cdot E \quad \text{Eq. 2}$$

$$D = e_s^T \cdot S + \varepsilon_s \cdot E \quad \text{Eq. 3}$$

where σ_m is the stress vector, D is the electric flux density vector, S is the strain vector, E is the electric field vector, c_E is the elasticity matrix (evaluated at constant electric field) and ε_s is the dielectric matrix (evaluated at constant mechanical strain).

The mechanical model for magnetostrictive material consists in a linear elastic model, fulfilled by the continuity equations on stationary case when the phase is subjected to maximal magnetostriction [7, 8, 16, 30]:

$$\nabla \cdot D = \rho_v \quad (\text{Gauss Law, Eq. 4})$$

$$\nabla \cdot \sigma = f_v \quad (\text{Cauchy momentum equation, Eq. 5})$$

where the divergence of the electrical displacement field, D , gives the free electric charge density, ρ_v , and the divergence of the stress tensor σ , gives the force per unit volume, f_v . The model also considers traction-free boundary conditions at outer surfaces and perfect bonding at all the interfaces of the structure (continuity equations of displacement and normal stress).

In order to obtain the electric potential (ϕ), the assumption of an electrode on the external surface of the ME structure is made, as an electrically floating electrode condition (where zero charge is held), therefore given by:

$$E = -\nabla \phi \quad \text{Eq. 6}$$

where E is the local electric field strength.

The obtained values for electric potential distribution were integrated over the electric potential interphase line, and α_{ME} is defined as (equation 7):

$$\alpha_{ME} = \left| \frac{dE}{dH} \right| = \left| \frac{\delta V}{d_{pzo}} \right| = \left| \frac{\int_{interface} \varphi \cdot dL}{d_{pzo} (cm) \cdot L} \right| \left(\frac{V}{cm} \right) \quad \text{Eq. 7}$$

where δV is the induced ME voltage, d_{pzo} is the piezoelectric thickness (in centimeters), φ is the electric potential, and L is the interface length.

In cases of experiments with more than one piezoelectric layer, the composite has to be treated as a device where piezoelectric layers are connected in series. The final ME coefficient, α_{ME} , is obtained by the sum of each ME coefficient, over each interface, as:

$$\alpha_{ME} = \sum_{INTERFACE} \alpha_{ME-INTERFACE} \quad \text{Eq. 8}$$

In other multilayer experiments, a unitary ME coefficient, called unitary α , or α_{unit} , can be defined by the sum of all partial electric potentials between magnetostrictive and piezoelectric layers, divided by the total piezoelectric length (in cm), as:

$$\alpha_{unit} = \left| \frac{dE}{dH} \right| = \left| \frac{\delta V}{d_{pzo}} \right| = \frac{\sum_{INTERFACE} \left| \int_{interface} \varphi \cdot dL \right|}{d_{pzo} (cm) \cdot L} \left(\frac{V}{cm} \right) \quad \text{Eq. 9}$$

where α_{unit} is the unitary ME coefficient for the multilayer, δV is the induced ME voltage, d_{pzo} is the total piezoelectric thickness (sum of all individual piezoelectric thicknesses, in cm), φ is the electric potential and L is the interface length. For symmetric cases, α_{unit} corresponds also to the average α_{ME} , or the sum of all α -values divided by the number of piezoelectric layers.

2.3 Multilayer ME laminate model experiments

ME performance of bilayered and multilayered laminates has been studied for piezoelectric ceramics and magnetostrictive metals [21, 22, 31, 32], but variations on the mechanical properties in the piezoelectric component (when including a piezoelectric polymer as PVDF, more flexible and with an amorphous semi-crystalline structure) may lead to variations in the ME performance of the composite. Finite element Method (FEM) simulation analyses were performed in order to evaluate the influence of the structural amorphous semicrystalline properties of PVDF on the ME performance of the ME laminates and to compare them with the results obtained for a ceramic piezoelectric material, as lead zirconate titanate (PZT-5H; $\text{Pb}[\text{Zr}_x\text{Ti}_{1-x}]\text{O}_3$; from now on PZT). A two-dimensional approximation was considered, establishing that the ME response of the structure will be constant along the width of the structure. Vitrovac 4040® was set to be the magnetostrictive part in the ME composite for both piezoelectric cases.

Piezoelectric calculations are based on linear models, as described by the constitutive equations, with the boundary condition of electrical ground in one of the surfaces or interphases in front of the other electrically isolated surface (interphase) where the electric potential was

evaluated. The applied deformation of the magnetostrictive material produces a deformation on the piezoelectric domain generating indirectly *via* strain an electric potential distribution on the interface between the piezoelectric and magnetostrictive material.

2.4 Magnetostrictive and piezoelectric material properties

For the piezoelectric component, PVDF was selected since it is the overall best piezoelectric polymer [5, 15]. Lead zirconate titanate PZT was selected as it is also among the most used piezoelectric ceramics.

Table 1: Structural and magnetostrictive properties of Vitrovac4040[25] and relevant electrical and mechanical properties of PVDF[33] and PZT-5H[16].

				0, 0, 0, 0.365, 0, 0, 0, 0, 0, 0.365}	
Electrical	Coupling Matrix	e	(C/m ²)	{0, 0, -4.761, 0, 0, -33.33}, {0, 0, 3.703, 0, 1.703, 0}, {1.703, 0, 0, 0, 0, 0}}	{0, 0, -6.62281, 0, 0, -6.62281}, {0, 0, 23.2403, 0, 17.0345, 0}, {17.0345, 0, 0, 0, 0, 0}}
	Relative Permittivity	ϵ_s	1	{{13, 0, 0}, {0, 13, 0}, {0, 0, 13}}	{{1704.4, 0, 0}, {0, 1704.4, 0}, {0, 0, 1433.6}}

2.5 Multi-layer configurations

Biphasic, triphasic and multiphasic ideal models (with perfectly bonded interphases) were modeled with different configurations and thicknesses for the two different piezoelectric materials (PVDF and PZT). The modelled configurations were classified according to their piezoelectric material, number of layers, and the type of external layer, into:

- M-P multi-layers: in this configuration the composite has an even number of layers with one magnetostrictive and one piezoelectric layer at the ends. The considered number of layers was 2, 4, 6, 8 and 10. The simplest case presented is the bi-layer PM configuration.
- M-M multi-layers: with an odd number of layers, the composite is always sandwiched between two magnetostrictive layers. The considered number of layers was 3, 5, 7 and 9. The simplest case presented is the trilayer MPM configuration.
- P-P multi-layers: with an odd number of layers, the composite is sandwiched between two piezoelectric layers. The considered number of layers was 3, 5, 7 and 11. The simplest case is the PMP trilayer configuration.

2.6 Independent thickness sweep experiment

First, independent magnetostrictive and piezoelectric thicknesses sweep were carried on. For the bilayer experiment, magnetostrictive and piezoelectric thickness were swept independently between 1 μ m and 1 mm. For both M-M and P-P configurations, the sweep included values for thickness between 10 μ m and 600 μ m. In all cases, when the magnetostrictive layer thickness was swept, the piezoelectric material thickness was held at a constant value of 110 μ m. Similarly, when the piezoelectric material thickness was varied, the magnetostrictive material thickness was set at a constant value of 25 μ m. For all cases, free and clamped configurations were analyzed. In the clamped configurations, the laminated structure was set to deform only along longitudinal direction in both bottom and top piezoelectric surfaces, similar to material fixed into a devices.

When more than one piezoelectric layer is included, the ME coefficient was obtained by equation 7, applied in each interface and then treated as a device where both piezoelectric layers are connected in series. Therefore, the final ME coefficient (α_{ME}) was obtained by the sum of each

ME coefficient, over each interface, as presented in equation 8. Then, the unitary ME coefficient, α_{unit} , was also calculated by equation after 9.

Figure 1 presents a schematic illustration of the simplest configuration of each experimental group, M-P, M-M and P-P.

Figure 1. Representative illustration of each type of simulation experiments, with the lowest considered number of layers. M-P bilayer on the left, MPM (M-M trilayer) configuration on the middle and PMP (P-P trilayer) configuration on the right.

In order to corroborate the trends obtained in the simulations, results were post-processed in order to be compared with data from the literature. For bilayers, Ryu *et al.* [22] reported a constant ME coefficient for higher Terfenol-D/PZT thickness ratios, that decays for thicknesses ratios below 10, establishing values for 0.5, 0.4 and 0.3 mm of PZT. On the case of MPM trilayers, Ryu *et al.* [23] established the theoretical expectation for the ME voltage coefficient (dE/dH) as function of the thickness ratio ($d_{\text{ms}}/d_{\text{pzo}}$) between Terfenol-D and PZT, showing that the output voltage increases while increasing the thickness ratio, saturating above $d_{\text{ms}}/d_{\text{pzo}}=10$. Peak values of α_{ME} as function of n-ratio ($2*d_{\text{ms}}/(2*d_{\text{ms}}+d_{\text{pzo}})$) were also reported for Metglas/PZT/Metglas [18]. These results were also confirmed experimentally for Terfenol-D/PZT/Terfenol-D laminates [23], for Co/PZT/Co heterostructures [34], and for the ME performance of different thickness ratio values of Terfenol-D/PMN-PT/Terfenol-D laminates [35].

2.7 Constant ME thickness multilayer experiment

In this case, the simulation experiments consisted in dividing a total thickness of $D_c=300 \mu\text{m}$ and $D_c=600 \mu\text{m}$ by the total number of layers of the composite. For example, for a number of layers $N=4$, each layer would be $75 \mu\text{m}$ thick for the $300 \mu\text{m}$ composite and $150 \mu\text{m}$ for the $600 \mu\text{m}$ composite. In order to assess the limitations of this structural analysis, results were compared with those obtained by Srinivasan [36, 37], that experimentally analyzed a $400 \mu\text{m}$ thick bilayer and multilayer thick-film structures of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO)-PZT and $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ (LCMO)-PZT. Those samples were fabricated with an equal number of manganite and PZT layers, between 2 and 8. The unitary ME coefficient was obtained from equation 9. The even-numbered layer experiment is schematically represented in Figure 2.

Figure 2: Schematic representation of M-P Multilayer ME simulation experiments, where n is the number of layers and the total thickness D_c was set to 300 μm and 600 μm .

3. Results and Discussion

3.1 Bi-layered composites

Figure 3 shows the simulated ME coefficient, α_{ME} , of the bilayer laminate structure for both PVDF and PZT as piezoelectric layers, including magnetostrictive (d_{ms}) and piezoelectric (d_{pzo}) thickness sweeps from 10 μm to 1 mm. The electric potential distribution of two particular simulations is also represented to illustrate the effect of the type of piezoelectric material.

Figure 3: Bilayered ideal laminate simulation results as a function of the magnetostrictive (d_{ms}) and piezoelectric (d_{pzo}) thickness. Figure (a) shows the ME coefficient, α (V/cm), of composites with constant piezoelectric thickness of (d_{pzo}) 110 μm , for different magnetostrictive material thicknesses (d_{ms}) and with constant $d_{\text{ms}}=25 \mu\text{m}$, for different piezoelectric thicknesses (d_{pzo}). Figure (b) and (c) shows the electric potential distribution for bilayered ME laminates with the following dimensions (b) $d_{\text{ms}}=90 \mu\text{m}$, $d_{\text{pzo}}=110 \mu\text{m}$ (c) $d_{\text{ms}}=25 \mu\text{m}$, $d_{\text{pzo}}=90 \mu\text{m}$, for both piezoelectric PVDF and PZT.

By establishing magnetostriction as a deformation, it may be expected the independence of the magnetostrictive layer thickness on the ME performance of the composite, which is confirmed by the results obtained for the magnetostrictive thickness sweep presented in Figure 3 (a) for bilayer composites. Small irregularities are introduced by the semicrystalline nature of PVDF in the ME response curves for magnetostrictive thickness sweep, resulting in above average α value for Vitrovac thicknesses of 20 μm and 300 μm , and below average α values for magnetostrictive thicknesses of 50 μm and 700 μm . For PZT thickness sweep (Figure xxx), it is obtained a constant ME coefficient for higher $d_{\text{ms}}/d_{\text{pzo}}$ thickness ratio, that decays for thicknesses ratios below 10, following the trends presented by Ryu *et al.* [22]. For PVDF-thicknesses below 100 μm also a constant ME response is obtained. On the other hand, irregular values are obtained for the ME response of samples with piezoelectric layer thickness above 200 μm , as shown in the Figure 3 (a).

Figure 3 (b) and (c) show the electric potential distribution for two cases of PVDF and PZT, revealing a strong influence of the PVDF's semi-crystalline constitution in the electrical properties. While the piezoceramic establishes a uniform laminar pattern of electric potential distribution in the polarization direction, the formation of clusters of heterogeneous regions with an irregular electric potential distribution is observed in the PVDF's simulation result. Therefore, this represents one of the main differences between the piezoelectric ceramic and the polymer, related to the irregular ordering in which dipoles generate the piezoelectric response in the PVDF's

samples, based on the random distribution of the crystalline phase, main responsible for the piezoelectric response, along the sample.

On average, for constant piezoelectric thicknesses, it is observed that the performance of the PVDF bilayered composite is about $\approx 12\%$ the one of the PZT counterpart. For equal piezoelectric thicknesses below $200\text{ }\mu\text{m}$ and constant magnetostrictive thickness of $d_{ms}=25\text{ }\mu\text{m}$, the PVDF composite shows an average α value of $\approx 8\%$ the one of the PZT counterpart.

3.2 Trilayered composites

Trilayered ME laminates may be composed of one piezoelectric layer sandwiched between two magnetostrictive layers (MPM configuration) or by one magnetostrictive layer intercalated between two piezoelectric layers (PMP configuration). When two piezoelectric layers are included, two sources of electric potential are involved which require considering the ME response α -values by equation (7 and 8). When configurations are to be compared, it is desirable to establish one single value that provides information on the performance of the material per unit piezoelectric length, which is obtained by equation (9), defining the unitary ME coefficient, α_{unit} .

Figure 4 and Figure 5 display the ME response of free and clamped, MPM and PMP ME laminates as compared to the bilayer configuration for magnetostrictive and piezoelectric thicknesses sweep, respectively, for each trilayered composite with PZT (a) and PVDF (b) as piezoelectric component. For the magnetostrictive sweep (Figure 4) of PZT trilayers, it is observed that the addition of one extra layer of magnetostrictive material (MPM) results in an increase of about 1 V/cmOe in the ME coefficient for all simulated magnetostrictive thickness range, when compared to the bilayer structure. It is also verified that the addition of an extra piezoelectric layer (PMP configuration) allows to double the ME response (by treating the composite as a single device and obtaining α_{ME} by eq. 9). The independence of α_{ME} over the magnetostrictive thickness remains generally valid for all configurations, with the only exceptions of specific PVDF configurations, in which the semicrystalline nature of PVDF plays a key role in establishing isolated peaks of ME response.

Figure 4: Simulation results of the ME coefficient of bilayered and trilayered ideal laminate composites as a function of the the magnetostrictive layer thicknesses (d_{ms}). Figure (a) shows the ME response (ME coefficient, α (V/cm)) of free and clamped, MPM and PMP ME structures with PZT as piezoelectric layer. Figure (b) shows the ME response (ME coefficient, α (V/cm)) of free and clamped, MPM and PMP ME structures with PVDF as piezoelectric material.

For MPM trilayered configurations with PZT as piezoelectric layer, clamping decreases the ME response progressively with decreasing magnetostrictive layer thicknesses (d_{ms}) below 150 μm , after which presented same order of α of the free trilayered structure. For clamped PVDF trilayer composites, these differences are even larger, leading to nearly zero ME response for magnetostrictive thickness of 70 μm for the MPM configuration. As PVDF is a viscoelastic material, and there is no structural damping considered, simulation results of the bilayer and the MPM-free configuration do not revealed relevant differences. The extra magnetostrictive layer has the effect of comprising (fixing, homogenizing) the ME coefficient to ≈ 11 V/cm, the same value as for the thinnest and thickest magnetostrictive thicknesses values in the bilayer configuration. For the same reason, the behaviour of the PVDF-PMP-clamped configuration for low thicknesses of Vitrovac surpassed the standard improvement of duplicating the ME performance of the bilayer (triplicating the α_{MEc} of the bilayer), decreasing α -values for higher magnetostrictive thicknesses. The lower performance was obtained for the particular case of Vitrovac with a thickness of 200 μm , the ME response increasing with increasing magnetostrictive layers thickness. For thinner magnetostrictive thicknesses, the PVDF clamped configuration show higher α -values than the free one, decreasing the performance for higher magnetostrictive thickness layers s and matching the performance of the free configuration for magnetostrictive thickness of 50 μm and later the bilayer performance for $d_{ms}=100$ μm . This allows to conclude that clamping would only be useful for thickness below $d_{ms}=50$ μm .

Figure 5: Bilayered and trilayered ideal laminate magneto-electric coefficient as a function of the magnetostrictive layer thickness (d_{pzo}). Figure (a) shows the ME response (ME coefficient, α (V/cm)) of free and clamped, MPM and PMP ME structures of PZT. Figure (b) shows the ME response (ME coefficient, α (V/cm)) of free and clamped, MPM and PMP ME structures of PVDF.

As shown in Figure 5, all PZT configurations present a constant α_{ME} value when the piezoelectric thickness is below 60 μm , decreasing for thicker piezoelectric thicknesses. Although the general trends for the three configurations remain similar, the clamped configuration exhibits the lowest ME performance, followed by the bilayered one, showing the free configuration the highest ME performance.

PVDF bilayer- and MPM free-configurations results displayed similar ME performance in the range of piezoelectric thickness below 200 μm , establishing that the free configuration shows a more stable trend than the bilayered one. The free configuration shows a stable α_{ME} of approximately 11 V/cm for the range of piezoelectric thickness below 200 μm , decreasing with increasing piezoelectric thicknesses above 200 μm . The clamped configuration shows a decaying value of α_{ME} for piezoelectric PVDF thickness in the range 10 to 40 μm , increasing for thicker PVDF layers, obtaining better ME performance than the bilayered and free-configurations for piezoelectric thicknesses above 200 μm .

While both, PZT and PVDF based free PMP configurations double the bilayer magneto-electric coefficient in the whole piezoelectric thickness range, the clamped configuration introduces some differences. When clamped, the PZT PMP configuration reduces ME performance when increasing the piezoelectric layer thickness. On the contrary, PVDF clamped configuration increases the ME response with increasing piezoelectric thickness, starting with a near-to zero response at 10 μm , but increasing the bilayer ME response for piezoelectric layer thickness of 20 μm and even being larger than the free configuration for piezoelectric thickness above 50 μm on each side of the Vitrovac layer.

This effect, as well as the exceptions where PVDF laminate's performance depended on the magnetostrictive thickness of the composite, can be explained by the results presented in Figure 6. Figure 6 shows that the electric potential in PZT laminates presents a laminar electric potential distribution on the polarization direction and homogeneous displacement field in the deformation

direction (y) through the thickness of the material. PVDF configurations, on the other hand, shows a non-homogeneous electric potential distribution. MPM-clamped configurations present an even less homogeneous electric potential distribution than the clamped ones, while PMP-clamped configurations present the opposite trend. These facts can be explained by the displacement field in the deformation direction that was 0.1 times the order of the free configuration for the MPM configuration, while presenting twice for the PMP counterpart. Therefore, obtaining higher reponse for clamped PMP configurations than for the free ones (for magnetostrictive thickness of $25\mu\text{m}$) and lower for the clamped MPM configurations than the free ones.

Figure 6: Representation of the electric potential distribution and vertical displacement of trilayered configurations. Figures (a) and (b) represent the electric potential distribution and displacement along the deformation direction of MPM laminates with $d_{\text{ms}}=25\mu\text{m}$ and d_{pzo} of $20\mu\text{m}$ (left), $50\mu\text{m}$ (middle) and $400\mu\text{m}$ (right), respectively. Figures (c) and (d) represent the

same information of PMP laminates with $d_{ms}=25\mu\text{m}$ and d_{pzo} of $30\mu\text{m}$ (left), $110\mu\text{m}$ (middle) and $400\mu\text{m}$ (right), respectively.

Ryu *et al.* [23] established the theoretical response for the ME voltage coefficient (dE/dH) of MPM heterostructures as function of the thickness ratio (d_{ms}/d_{pzo}), presenting that the output voltage increases while increasing the thickness ratio, saturating above $d_{ms}/d_{pzo}=10$. This trend is corroborated in measurements in [23] Terfenol-D/PZT/Terfenol-D laminates, [18] Metglas/PZT/Metglas configurations, [34] Co/PZT/Co heterostructures and [35] for the ME performance of different thickness ratio values of Terfenol-D/PMN-PT/Terfenol-D laminates. Wong [35] also measured α_{ME} as function of bias magnetic field for PMP laminates based on PMN-PT/Terfenol-D/PMN-PT with three different thickness ratio of 0.4, 1.4 and 2.4. The results showed that the ME performance of the PMP configurations increases with increasing the thickness ratio. Finally, a comparison was established between the ME response of MPM and PMP composites, where both configurations rapidly increased their unitary ME behaviour (α_{unit}) with increasing the thickness ratio, until reaching saturation. The only difference observed was that MPM configurations reached saturation with a higher slope than PMP configurations, and with a higher ME coefficient (PMP- α in saturation is approximately an 85% of MPM- α).

Figure 7 shows a comparison between the ME response of MPM and PMP composites with PZT (a) and PVDF (b) as piezoelectric layers.

Figure 7: Theoretical ME response for PMP and MPM configurations as a function of magnetostrictive/piezoelectric thickness ratio (d_{ms}/d_{pzt}) for PZT-trilayers (a) and PVDF-trilayers (b).

While the trends for PZT configurations confirm the ones established by Ryu *et al.* [23], they show a much more abrupt slope for MPM configurations. PZT-PMP laminates ME response as a function of thickness ratio shows an increasing ME response for thickness ratio below 0.3, and saturating above this thickness ratio value. PVDF-PMP laminate simulations show a more disordered trend, with a decreasing α for thickness ratios below 0.1, then increasing until reaching saturation for a thickness ratio of 0.3. This difference is due to the mechanical properties of PVDF, that do not allow to completely transmit deformations when the piezoelectric thickness is too large, therefore decreasing the ME response.

While the trends presented by Wong[35] are confirmed by PZT MPM and PMP composites simulation results, PVDF MPM and PMP simulations results do not lead to clear tendencies. PZT MPM and PMP simulation results display high-sloped increase of the ME response with increasing thickness ratio, the MPM-configuration reaching saturation for a smaller thickness ratio than the PMP configuration, with a higher ME coefficient in about 2 V/cm. On the other hand, PVDF MPM and PMP configurations, present curves that match the high-sloped increase of the ME response with increasing thickness ratio and saturating afterwards, but the higher viscoelasticity of PVDF plays an essential role and, after unity of thickness ratio is reached, PMP configurations lead to a better unitary ME response than MPM configurations.

3.3 Multi-layered composites

Figure 8 shows the simulation results for M-M and P-P configurations ME response as function of the thickness ratio (d_{ms}/d_{pzo}) for PVDF and PZT piezoelectric layers.

PZT M-M and P-P laminates show an enhanced and constant value ME response for either undersized magnetostrictive thicknesses or bulky piezoelectric thicknesses, as presented in Figure 8 (a) and Figure 8 (c). It is shown, that the ME coefficient α_{unit} increases rapidly while increasing thickness ratio for both configurations, saturating around a thickness ratio of 0.3 and showing therefore a constant response for higher thickness ratios. For high thickness ratios the M-M configuration shows a higher stable ME value on about 1 V/cm, showing a slight improvement by the addition of more layers on the M-M configuration. With respect to the P-P configuration, on the other hand, the highest ME coefficient is presented by the 5-Layer P-P configuration. For both M-M and P-P configurations and low thickness ratio (below 0.2, meaning high piezoelectric thicknesses and low magnetostrictive thicknesses), the highest number of layers present to introduce an increase on the ME performance of the material is XXXXX.

Figure 8: Simulation results for MM and PP multilayered configurations (L indicating the number of layers) as a function of the thickness ratio ($d_{\text{ms}}/d_{\text{pzo}}$), for PZT M-M (a), PVDF M-M (b), PZT P-P (c) and PVDF P-P configurations.

PVDF M-M and P-P configurations also preserve ME drifts over magnetostrictive thickness, in particular for low-ordered multilayer configurations (below 5 layers). For higher number of layers, the ME response is governed by the malleability and semicrystalline behavior of the piezoelectric layer, therefore including high number of nonlinearities (peaks). This fact indicates a better ME response when avoiding damping and magnetic losses in the coupling factor. When a high number of layers are involved in the composition of the laminate, the mechanical effects of the viscoelastic PVDF lead to more heterogeneous patterns of ME coefficient. Although preserving the trend reported in [35] for low layer number in PVDF P-P multilayered configurations, the introduction of higher number of PVDF layers in the composite induces a nonlinear behavior. Those nonlinearities present themselves as peak values for specific thickness ratios, establishing improvements when damping or magnetic losses are not considered. It can be also noticed, that the trend of PVDF-PMP α_{ME} versus thickness ratio is preserved for the 5-layered P-P configuration, obtaining high ME response values for thickness ratios below 0.1, then decreasing quickly to increase again and saturating for thickness ratios above 0.6, even achieving similar ME performance values.

Figure 9 shows simulation results for particular cases of symmetric multilayered M-M and P-P configurations with unity of thickness ratio ($d_{\text{ms}}=d_{\text{pzo}}$) for thicknesses of 25 μm (25/25) and 110 μm (110/110).

PZT M-M and P-P configurations present higher ME coefficient for lower thicknesses, with higher values for M-M than P-P configurations. M-M configurations show a uniform ME coefficient as a function of the number of layers, with higher values than the bilayer composite. P-P configurations, on the other hand, show the highest ME response for the 5-Layer configuration, reaching the value of the M-M configuration for the thin composite (25/25 P-P).

Figure 9: Simulation results for symmetric M-M and P-P multilayered configurations as function of the number of layers, N , when magnetostrictive and piezoelectric layers have the same thickness, of 25 and 110 μm , for M-M PZT/Vitrovac (a), M-M PVDF/Vitrovac (b), P-P PZT/Vitrovac (c), and P-P PVDF/Vitrovac (d).

PVDF M-M and P-P configurations are influenced by the viscoelasticity of the electroactive polymer. M-M configurations show clear trends of decreasing ME coefficient with increasing number of layers, presenting a maximum performance for the bilayer structure. P-P configurations show no stable behavior, exhibiting a ME coefficient 4 times higher than any other configuration for the 25 μm -7 layer composite, of 48 V/cm. Figure 10 shows the electric potential distribution for 3, 5, 7 and 11 layered ME composites with 25 μm layer thickness. It is observed in (c) that the top piezoelectric layer of the 7-layerd composite is producing under deformation a series of dipolarities that are at the origin of this enhanced ME response. Beside this isolated enhanced ME responses of the 7-layered 25/25 ME composites, results show better performance for P-P configurations of 110/110 and 7 or 9 layers, improving the ME response for the latter around 3 V/cm in comparison with the bilayer configuration.

Figure 10: Electric potential distribution of P-P configurations with 3 (a), 5 (b), 7 (c) and 11 (d) layers with a colour range between 2V (dark red) and -2V (dark blue)

3.4 Constant ME thickness multilayer experiment

Srinivasan *et al.* [37] evaluated the ME effect in bilayers and multilayers of thick-film structures composed of (LSMO)-PZT and (LCMO)-PZT, in which samples were fabricated with an equal number of piezoelectric and magnetostrictive layers, between 2 and 8, maintaining constant the total thickness, D_C , of the device and the thickness of both magnetostrictive and piezoelectric phases. This study concluded that transverse ME voltage is lower in multilayers compared to bilayers, and that the relation is linear and proportional to the number of layers, N . Simulation results for PZT and PVDF laminates are shown in Figure 11 (a) and (b), respectively, with corresponding electric potential distribution Figure 11 (c).

Figure 11: Simulations of peak ME voltage coefficients for PZT M-P (a) and PVDF M-P (b) multilayers as a function of the numbers of layers, N . In figure (c) it is shown the electric potential distribution for FEM simulations with PZT (top) and PVDF (bottom) M-P multilayers, with 2 to 10 layers, with the cases of 600 μm and 300 μm of total thickness.

PZT M-P laminates show an increase in the ME response when divided into multilayers for both total thicknesses, D_c , of 300 and 600 μm , a different trend that the one reported in [37]. The differences on the simulations based on PZT as piezoelectric layers can be related to the lack of consideration, in the present simulations, of structural damping and magnetic losses, which may play a relevant role when more layers are introduced. Although the 300 μm -thick PVDF M-P laminate shows the same results as the PZT M-P simulation trends, the PVDF multilayer of total thickness of 600 μm follows Srinivasan's *et al.* results, showing a slightly improved ME unitary performance as a lower number of layers is involved. The electric potential distribution in Figure 11 (c) shows the homogeneous pattern in the PZT configurations, and the inhomogeneous patterns for PVDF M-P laminates. While the thinner PVDF M-P ME composites already presented a dipolar inhomogeneous pattern in the bilayer configuration, the composites with total thickness of 600 μm , only show this effect in the top of the piezoelectric layer for $N=6$ or lower. In these cases, the poles appeared to be bigger and therefore the ME performance is enhanced, even showing an approximately homogeneous electric potential distribution pattern for at least one of the piezoelectric internal layers for $N=4$ and $N=6$.

Figure 12 presents the results of the ME unitary coefficient, α_{unit} , as function of the configuration (M-M, P-P or MP) and de number of piezoelectric layers in the composite, when the total thickness is held constant at 300 and 600 μ m.

Figure 12: Unitary ME coefficient for PZT MP, M-M and P-P (a) and PVDF MP, M-M and P-P (b) constant thickness composites with a total thickness of 300 μ m and 600 μ m as function of the number of piezoelectric layers, N_{pzo} .

PZT M-M laminates show increasing ME response with increasing the number of piezoelectric layers (N_{pzo}), with a higher α_{ME} for thinner layers and low amount of layers but reaching similar results for more than 7 layers composites ($N_{pzo} > 3$). Those results are similar to ones for MPM laminates. PZT P-P experiments also show higher α_{unit} for thinner layers, taking approximately a constant value. In general, all PZT configurations show the trend of increasing ME response with increasing the number of piezoelectric layers, obtaining an enhanced response for M-M configurations with less than 4 piezoelectric layers, but reaching its maximum value for the P-P configurations with 4 electroactive phases. It is to notice, that these higher values for high number of layers can decrease when considering other effects as coupling parameters for bonding properties or magnetic losses. It can be concluded than in M-P and P-P configurations the role of the composite's thickness plays a larger role than in M-M configurations. These showing more stable values, with a maximal difference of 1V/cm for 1 piezoelectric layer, for low number of piezoelectric layers.

Based on the elasticity and semicrystallinity of the piezoelectric polymer, PVDF M-M configurations show larger α -values for low-layered composites with higher thickness, and with more stable unitary α over different number of layers (with an almost constant value of unitary α , of approx. 10.7-10.9 V/cm). For 5 or more layers ($N_{pzo} > 2$), PVDF M-M experiments show a clear trend of maintaining α -values independently of the total thickness of the composite. As obtained for the PVDF M-P constant thickness configurations, PVDF M-M composites performed better as a bulk-bilayered composite than in a thinner multi-layered ones. Also considering the results obtained by Srinivasan *et al.* [36] and considering damping, bonding and magnetic losses, it may be concluded that thicker bilayered PVDF composites will result in an enhanced performance over multilayered thinner ones.

M-P configurations, on the other hand, present high ME response for a low number of layers and higher thicknesses, and an increasing ME response of thin composites with the amount of layers. While the bilayered M-P 600 μm configuration shows the highest α , of approx. 12.4 V/cm, the bilayered M-P 300 μm shows the lowest value of 7 V/cm. P-P configurations, on the other hand, show no clear trends, as the highest achieved value is established by the P-P 600 μm with more than double (27 V/cm) the highest M-P ME coefficient value. This confirms also, that P-P configurations may not presented clear trends and that the elasticity and morphology of PVDF has a larger influence in this configuration.

4. Conclusions

A structural simulation model based on Finite Element Methods (FEM) for magnetoelectric (ME) composites was developed in order to establish the influence of the mechanical properties and morphology of the different layers in the ME response. Ceramic (PZT) and polymer (PVDF) based magnetostrictive multilayered composites were analyzed according to their configuration, thickness and number of layers. This comparison, allows to establish the influence of the structural properties of the piezoelectric layers in the ME coupling, leading to a better understanding of composites behavior.

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PZT composites simulations allowed to obtain clear trends of enhanced composites, for bilayers with thinner piezoelectric thicknesses (below 200 μm) and magnetostrictive thicknesses above 50-60 μm for clamped configurations. Good agreement was established when comparing with the trends reported experimentally in the literature for magnetostrictive to piezoelectric thickness ratio of multilayers, presenting a curve that rapidly increases their α_{unit} with increasing thickness ratio until $n=0.3$, when saturation is reached. The difference with M-M configurations is that they reached saturation with a higher slope than P-P configurations, to a higher value, therefore presenting an optimal configuration for multilayers with external magnetostrictive phases (M-M configurations), with thickness ratio above 0.2 (being the thickness of the magnetostrictive phase higher than 5 times the electroactive phase's thickness), of about 86.7 V/cm.

PVDF bi-layers, on the other hand, exhibit a constant α_{ME} values of 10.71 V/cm, for polymer layers below 110 μm , and then some higher and lower α_{ME} values. This established a clear influence of the semicrystalline nature and viscoelasticity of PVDF in this heterogeneous ME behaviour with isolated peak values. These effects introduce difficulties in establishing trends on the influence of thicknesses and configuration on the ME coefficient of the multi-layered composites. It may be established that M-M configurations show more stable behaviour and trends over different number of layers, while P-P configurations present regions with peaks that went off the expected trend. It can be also established that M-P configurations are severely dependent on the composite's thickness. For configuration with less than 3 piezoelectric layers trends show no particular influence of magnetostrictive thickness over the ME performance of free PVDF multi-layered configurations, and a slight decrease of α_{ME} above piezoelectric thicknesses of 150 μm . Clamped configurations, on the other hand, show a ME performance that is dependent of magnetostrictive and piezoelectric thicknesses. For free configurations, thin PVDF thicknesses improve the ME response of the composite, with the possibility of the incorporation of more than one layer to enhance the performance of the structure.

As PZT-based ME composites present clear trends, it appears necessary to incorporate bonding effects, damping and magnetic losses to the reported simulations. Bonding and damping

effects will be reinforced with the incorporation of more layers in the composite and the present structural model should be improved by incorporating these variables into simulations. With the incorporation of damping and bonding effects, multi-layer structural simulation results would be much more effective and would allow a better understanding of the peak variations presented by P-P configuration. Also, this analysis suggested that every configuration of PVDF-laminate has to be carefully analyzed within the mounting conditions. Resources should be devoted into establishing a nonlinear magnetostrictive model that could be coupled to this structural model, in order to obtain more information about the magnetostrictive performance of the material, and to compare different options of magnetostrictive materials. PVDF's biocompatibility, elasticity and malleability, makes it suitable for a high number of technological applications.

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