MICROMECHANICS MODELING OF MAGNETOELECTRIC COMPOSITES

Tomas I. Muchenik, Ever J. Barbero
West Virginia University
395 Evansdale Dr.
Morgantown, WV, 26505

ABSTRACT

A novel model is proposed to optimize the design of magnetoelectric sensor composites. The influence of the properties of piezoelectric and piezomagnetic materials, volume fraction, and magnetic field orientation on the output magnetoelectric voltage are evaluated. Comparison of model results with experimental data and prior work is presented.

1. INTRODUCTION

The magnetoelectric effect appears as a result of the coupling between magnetic and electric fields. This effect can be obtained in single phase materials and in composite materials. Single phase materials are limited by their operational temperature, which is bellow room temperature, and their small magnetoelectric coupling [1] [2]. Magnetoelectric composites overcame these limitations. A magnetoelectric composite relies on the "product property" (i.e., coupling) between piezoelectric (PE) and piezomagnetic (PM) phases [3]. These composites can be used to generate magnetoelectric behavior from materials which do not possess inherent magnetoelectric effect. When a magnetic field is applied to the composite, it will produce a strain in the PM phase which is mechanically transferred to the PE material, which will then produce an electric voltage.

Traditional magnetic sensors, like Hall or magnetoresistive sensors, require a power supply. Since in magnetoelectric composites the electric signal is spontaneously produced as a result of the applied magnetic field, a power supply is not required [4]. These magnetoelectric sensors have proven picotesla sensitivities. This sensitivity is the result of on high magnetoelectric voltage coupling, which is the voltage (electric field) produced in the composite when a magnetic field is present. In this way, the magnetoelectric composite becomes a sensitive detector of changes in magnetic field without requiring a power supply. Thus, the device can be very small and portable. Applications include detection of improvised explosive devices (IED), and so on.

To achive high magnetoelectric coupling, several materials have been used as PM phase: ferrites, Terfenol-D and Metglas, and PE phase: PZT, PVDF, PZN-PT and BaTiO [4] [5] [6] [7] [8]. The influence of constituent material properties on magnetoelectric voltage (sensitivity) is not easily discerned without a model. Therefore, further study is needed to design a composite structure that will have the optimum sensitivity among the available constitutive materials. Additionally, the PM and PE volume fractions, as well as the

direction in which the magnetic field is applied to the composite, have strong influence on the sensitivity of the device.

2. MODEL

The analytical model is based in the constitutive equations proposed by Osaretin *et al.* [9]:

$$s = S\,\sigma + d\,E + q\,H$$

$$D = d\,\sigma + \epsilon\,E \tag{1}$$

$$B = q\,\sigma + \mu\,H$$

where s is the strain tensor, σ is the stress tensor, E is the electric field vector, H is the magnetic field vector, D is the electrical displacement vector, B is the magnetic induction vector, S is the compliance matrix, d is the PE constitutive matrix, q is the PM constitutive matrix, ϵ is the electric permittivity matrix and μ is the magnetic permeability matrix. These equations describe the behavior of the PM and the PE phases.

Two different setups are used (Figure 1): transverse electric polarization with transverse magnetic polarization (TT), and transverse electric polarization with longitudinal magnetic polarization (TL). The PE phase is most commonly polarized in the transverse direction. Otherwise, an insulator is needed at the interface to prevent charge leakage from the PE through the PM phase, since the latter is highly conductive.

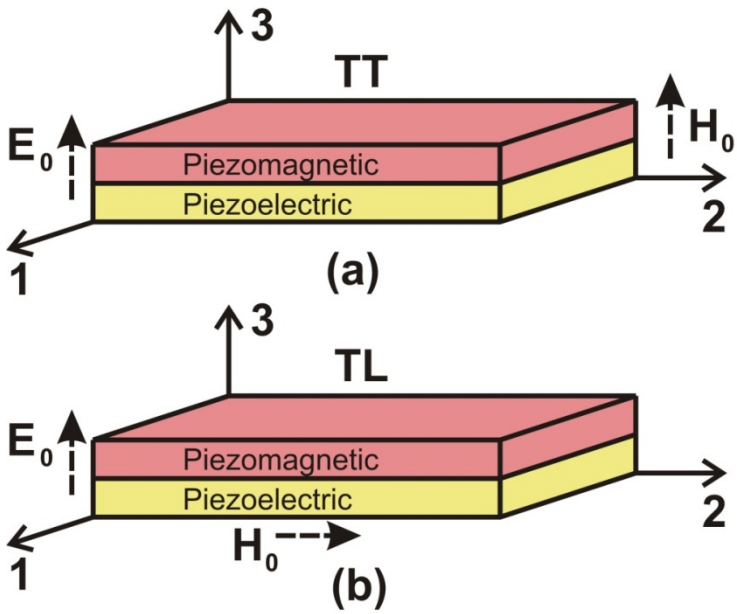

Figure 1: Geometry of the magnetoelectric laminate composite modeled with two different configurations: (a) Transverse electric polarization and transverse magnetic polarization (TT) and (b) Transverse electric polarization and longitudinal electric polarization (TL).

There two possible geometric configurations shown in Figure 1 (a) and (b) are implemented by appropriate boundary conditions, as follows.

2.1 Transverse Electric and Magnetic Polarization (TT)

Using Equation (1) for the first term of the strain vector (s_1), the following equations are obtained:

$$\begin{aligned} s_1 &= S_{11}\,\sigma_1 + S_{12}\,\sigma_2 + S_{13}\,\sigma_3 + d_{31}\,E_3 + q_{31}\,H_3 \\ &= \sigma_1(S_{11} + S_{12}) + d_{31} + q_{31}\,H_3 \end{aligned} \tag{2}$$

The following boundary conditions are applied: $\sigma_1 = \sigma_2$ because of the symmetry of the applied fields and geometry, $\sigma_3 = 0$ since the laminate is not restrained to deform in the 3-direction. The terms of the PE and PM coupling consider the polarization in the 3-direction shown in Figure 1. This constitutive equation is valid for both PM and PE phases. Since $d_{31}^{PM} = 0$ in the PM phase, (2) reduces to:

$$s_1^{PM} = \sigma_1^{PM}(S_{11}^{PM} + S_{12}^{PM}) + q_{31}^{PM}\,H_3^{PM} \tag{3}$$

Since $q_{31}^{PE} = 0$ in the PE phase, (2) reduces to:

$$s_1^{PE} = \sigma_1^{PE}(S_{11}^{PE} + S_{12}^{PE}) + d_{31}^{PE}\,H_3^{PE} \tag{4}$$

From Equation (1), the electric displacement in the 3-direction, D_3, with $d_{31} = d_{32}$, is expressed as:

$$D_3 = 2\,d_{31}\sigma_1 + \epsilon_{33}E_3$$

Since d $= 0$ in the PM phase,

$$D_3^{PM} = \epsilon_{33}^{PM}E_3^{PM} \tag{5}$$

And in the PE phase:

$$D_3^{PE} = 2\,d_{31}^{PE}\,\sigma_1^{PE} + \epsilon_{33}^{PE}\,E_3^{PE} \tag{6}$$

The same procedure is used to obtain the magnetic flux in the 3-direction for the PM phase:

$$B_3^{PM} = 2\,q_{31}^{PM}\,\sigma_1^{PM} + \mu_{33}^{PM}\,H_3^{PM} \tag{7}$$

And for the PE phase:

$$B_3^{PE} = \mu_{33}^{PE}\,H_3^{PE} \tag{8}$$

Additional boundary conditions are needed to solve this problem. Both layers are bonded, meaning that the deformation and strain on both layers are equal:

$$s_1^{PE} = s_1^{PM} \tag{9}$$

By force equilibrium, the force in the PM and PE phases have the same magnitude and opposite direction. The PM volume fraction, χ, is defined to take into account laminates with different thicknesses:

$$\chi = \frac{V^{PM}}{V^{PM} + V^{PE}} \tag{10}$$

where V is the volume. With this definition of χ, the equilibrium of forces is expressed as:

$$\sigma_1^{\mathrm{PM}} = -\sigma_1^{\mathrm{PE}} \left(\frac{1}{\chi} - 1\right) \tag{11}$$

The electric displacement, D, is the same in the PE and PM phases since the current is the same in both laminas as it runs between the top and the bottom of the laminate,

$$D_3^{PM} = D_3^{PE} \tag{12}$$

The same applies to the magnetic flux:

$$B_3^{PM} = B_3^{PE} \tag{13}$$

It is convenient to define the averaged magnetic and electric field since these are the values that are measured or applied to the composite:

$$\begin{aligned} E^{avg} &= E_3^{PE}\,(1-\chi) + E_3^{PM} \\ H^{avg} &= H_3^{PE}\,(1-\chi) + H_3^{PM} \end{aligned} \tag{14}$$

The value of H^{avg} has to be fixed to calculate the magnetoelectric response, but since the material has a linear response to the applied magnetic field, the magnetoelectric response is independent of the value chosen. A magnetic field $H^{avg} = 1\ A/m$ was selected for the calculations.

The last boundary condition is related to the magnetoelectric coefficient that will be measured. If the magnetoelectric voltage coefficient is measured, the electric displacement is zero ($D_3 = 0$). If the magnetoelectric charge coefficient is measured, the electric field is zero ($E^{avg} = 0$). The system of equations 3-14 is solved using Mathematica [10]. Finally, the magnetoelectric voltage is calculated as follows:

$$ME\ voltage = \frac{E^{avg}}{H^{avg}} \tag{15}$$

2.2 Transverse Electric and Longitudinal Magnetic Polarization (TL)

A similar approach can be used to solve the constitutive equations for the TL configuration. Using Equation 1 for the first and second terms of the strain vector (s_1, s_2), the following equations are obtained:

$$\begin{aligned} s_1 &= S_{11}\,\sigma_1 + S_{12}\sigma_2 + S_{13}\,\sigma_3 + d_{31}\,E_3 + q_{31}\,H_2 \\ s_2 &= S_{12}\,\sigma_1 + S_{11}\,\sigma_2 + S_{13}\,\sigma_3 + d_{31}\,E_3 + q_{33}\,H_2 \end{aligned} \tag{16}$$

In this case $\sigma_1 \neq \sigma_2$ since the magnetic field will produce an expansion in the 2-direction and a contraction in the 1-direction. The stress in the 3-direction is zero ($\sigma_3 = 0$) since the laminate is not restrained to deform in the 3-direction. The terms of the PE and PM coupling consider the polarization in the 3-direction and 2-direction shown in Figure 1. This constitutive equation is valid for both the PM and PE phases. For the PM phase ($d_{31}^{PM} = 0$), it reduces to:

$$\begin{aligned} s_1^{PM} &= S_{11}^{PM}\,\sigma_1^{PM} + S_{12}^{PM}\,\sigma_2^{PM} + q_{31}^{PM}\,H_2^{PM} \\ s_2^{PM} &= S_{12}^{PM}\,\sigma_1^{PM} + S_{11}^{PM}\,\sigma_2^{PM} + q_{33}^{PM}\,H_2^{PM} \end{aligned} \tag{17}$$

For the case of the PE phase ($q_{31}^{PE} = q_{33}^{PE} = 0$):

$$\begin{aligned} \mathrm{s_1^{PE}} &= \mathrm{S_{11}^{PE}\,\sigma_1^{PE} + S_{12}^{PE}\,\sigma_2^{PE} + d_{31}^{PE}\,E_3^{PE}} \\ \mathrm{s_2^{PE}} &= \mathrm{S_{12}^{PE}\,\sigma_1^{PE} + S_{11}^{PE}\,\sigma_2^{PE} + d_{31}^{PE}\,E_3^{PE}} \end{aligned} \tag{18}$$

From Equation 1, the electric displacement in the 3-direction, D_3, with $d_{32} = d_{31}$, $\sigma_3 = 0$, is expressed as:

$$\mathrm{D_3 = d_{31}\,\sigma_1 + d_{31}\,\sigma_2 + \epsilon_{33}\,E_3} \tag{19}$$

When this equation is applied to the PM phase, the following equation is obtained:

$$\mathrm{D_3^{PM} = \epsilon_{33}^{PM}\,E_3^{PM}} \tag{20}$$

For the PE phase:

$$\mathrm{D_3^{PE} = d_{31}^{PE}\,\sigma_1^{PE} + d_{31}^{PE}\,\sigma_2^{PE} + \epsilon_{33}^{PE}\,E_3^{PE}} \tag{21}$$

The same procedure is used to obtain the magnetic flux in the 2-direction for the PM phase:

$$\mathrm{B_2^{PM} = q_{31}^{PM}\,\sigma_1^{PM} + q_{33}^{PM}\,\sigma_2^{PM} + \mu_{33}^{PM}\,H_2^{PM}} \tag{22}$$

For the PE phase:

$$\mathrm{B_2^{PE} = \mu_{33}^{PE}\,H_2^{PE}} \tag{23}$$

Since the PM and PE layers are bonded, the deformation and strain on both layers are equal:

$$\begin{aligned} \mathrm{s}_1^{\mathrm{PE}} &= \mathrm{s}_1^{\mathrm{PM}} \\ s_2^{PE} &= s_2^{PM} \end{aligned} \tag{24}$$

By force equilibrium, the force produced by the PM phase and resisted by PE phase has the same magnitude and opposite directions:

$$\begin{aligned} \sigma_1^{\mathrm{PM}} &= -\sigma_1^{\mathrm{PE}} \left(\frac{1}{\chi} - 1\right) \\ \sigma_2^{\mathrm{PM}} &= -\sigma_2^{\mathrm{PE}} \left(\frac{1}{\chi} - 1\right) \end{aligned} \tag{25}$$

The electric displacement is the same in the PE and the PM phases since the current is produced and it flows between the top and the bottom of the laminate:

$$\mathrm{D}_3^{\mathrm{PM}} = \mathrm{D}_3^{\mathrm{PE}} \tag{26}$$

Since the magnetic field is applied in the 2-direction, the magnetic field is the same in both phases:

$$\mathrm{H}_2^{\mathrm{PM}} = \mathrm{H}_2^{\mathrm{PE}} = H_2 \tag{27}$$

Again, it is convenient to define the average electric field, since this is the value that is externally measured:

$$\mathrm{E}^{\mathrm{avg}} = \mathrm{E}_3^{\mathrm{PE}} (1 - \chi) + \mathrm{E}_3^{\mathrm{PM}} \chi \tag{28}$$

These equations are solved using Mathematica [10].

2.3 Materials Properties

There are several PE and PM materials that can be combined to achieve a magnetoelectric composite. In this work CFO (Co_2FeO_4), Terfenol-D, and Metglass were selected as PM materials. The mechanical, magnetic and electric properties are given in Table 1. The PE materials considered are: PZT ($Pb(Zr,Ti)O3$), PVDF (Polyvinylidene fluoride), PZN-PT ($Pb(Zn_{0.3}Nb_{0.7})O_3\ PbTiO_3$) and BaTiO ($BaTiO_3$). The properties of the PE materials are shown in Table 2.

Property	$CoFe_2O_4$(CFO)	Terfenol-D	Metglas
$S_{11}(10^{-12}/Pa)$	6.5	33.3	8.5
$S_{12}(10^{-12}/Pa)$	-2.4	-10	-2.5
$q_{31}(10^{-12}m/A)$	-566	-4729	-5800
$q_{33}(10^{-12}m/A)$	1880	15707	12000
μ/μ_0	2	2	2
ϵ/ϵ_0	1	1	1

Table 1: Mechanical, electrical, and magnetic properties of selected PM materials obtained from [8], [9] and [11].

Property	PZT	PVDF	PZN-PT	BaTiO
$S_{11}(10^{-12}/Pa)$	16.5	420	6.9	9.03
$S_{12}(10^{-12}/Pa)$	-4.78	-198	-2.1	-3.21
$d_{31}(10^{-12}C/N)$	-274	-21	-2800	-78
ϵ/ϵ_0	3400	10	5500	1345
μ/μ_0	1	1	1	1

Table 2: Mechanical, electrical, and magnetic properties of selected PE materials obtained from [8], [9] and [11].

3. RESULTS

3.1 Model Validation

The magnetoelectric voltage coefficient calculated in this work was compared to the results reported by Osaretin *et al.* and Bichurin *et al.* and the experimental data reported by Harshe *et al.* [9] [11] [12]. These results can be seen in Figure 2. The results obtained in this work are in agreement with previously reported results and experimental data. The data shows a tradeoff between the amount of PE and PM phase.

When higher amounts of PM phase are used (higher PM volume fraction χ), it gives higher deformation resulting in a higher electric field in the PE phase. The relationship between the electric field in the PE phase and the electric field in the composite can be seen in Equation 14, showing that the total electric field depends on the electric field produced in the PE phase and the PM volume fractions (or thicknesses).

For PM volume fraction below 1/2, increasing the thickness of the PM phase gives an increase of the total electric field, since the increase of the electric field in the PE phase is

higher than the decrease of the thickness of the PE phase. But in the case of PM volume fraction above 1/2, even though the electric field in the PE phase increases, the decrease in PE phase thickness results in a decrease of the total electric field of the composite. Thus the parabolic response observed in Figure 2.

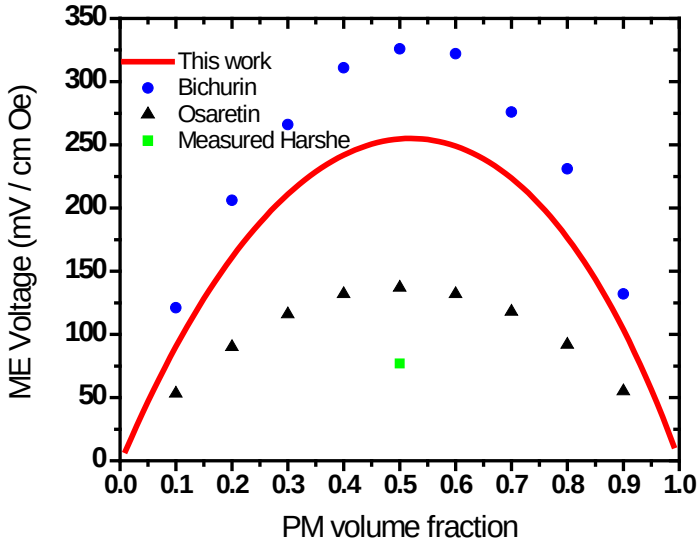

Figure 2: Magnetoelectric voltage coefficient for CFO and PZT composite for different volume fractions (χ) compared with model proposed by Bichurin *et al.*, and Osaretin *et al.* and measured values by Harshe *et al.* [9] [11] [12].

3.2 Magnetoelectric Voltage for TT and TL Configurations

As it can be seen in Figure 1, the TT and TL configurations differ in the orientation of the magnetic field being transverse (out of plane) and longitudinal (in plane), respectively. In order to compare the magnetoelectric voltage coefficient, a composite made of Metglas and PZN-PT is modeled in both configurations.

The results can be seen in Figure 3. For the TT configuration, the maximum magnetoelectric voltage is 1.6 V/A for a PM volume fraction of 38%. For the TL configuration, the maximum magnetoelectric voltage is 13 V/A for a PM volume fraction of 47%. The higher magnetoelectric coupling for the TL configuration is mainly due to the fact that the PM coupling in the 13-direction (q_{13}) is three times smaller than the PM coupling in the 33-direction (q_{33}). The other combinations of PM and PE materials show similar behavior since the PM couplings follow the same relationship in all the cases. This indicates that the optimum magnetoelectric coupling is obtained with the TL configuration.

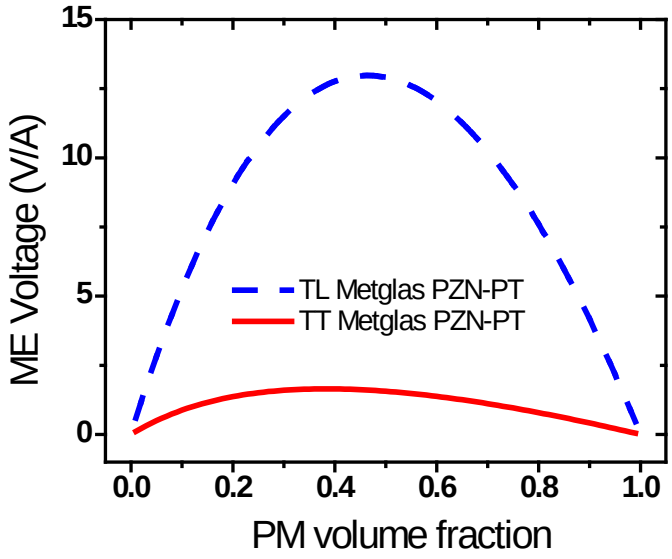

Figure 3: Magnetoelectric voltage for Metglas PZN-PT laminate composite modeled with two different configurations: (a) Transverse electric polarization and transverse magnetic polarization (TT) and (b) Transverse electric polarization and longitudinal electric polarization (TL).

3.3 Influence of Mechanic, Electric, and Magnetic Properties on Magnetoelectric Voltage

The magnetoelectric coupling in composites structures is highly dependent on the mechanical, magnetic, and electric properties of its constituents. In order to analyze the influence of the PE coupling (d_{31}) of PZN-PT on the magnetoelectric voltage, two values of d_{31} are used: half the PE coupling and two times the nominal PE coupling ($1400\ and\ 5600\ 10^{-12}C/N$).

The results can be seen in Figure 4. The optimum volume fraction is the same for all the cases (47%), indicating that the compromise between the electric field produced and the thickness is not influenced by the PE coupling. The maximum magnetoelectric voltage for the case using half and twice the original value of the PE constant was 6.9 V/A and 27.9 V/A, respectively. This indicates that the magnetoelectric voltage is linearly related to the PE coupling. The PM coupling has the same influence on the magnetoelectric voltage.

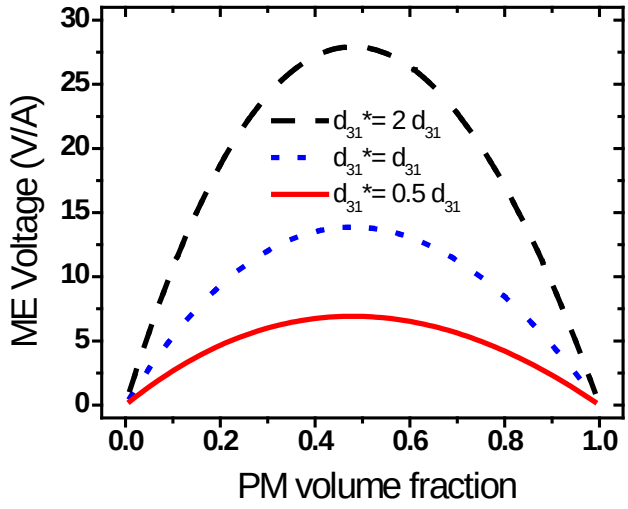

Figure 4: Magnetoelectric coupling for PZN-PT Metglas composite using different values for the PE coupling.

Another important electrical property is the dielectric constant (ϵ) of the PE phase. The influence on the magnetoelectric voltage is evaluated by changing the value of ϵ in the PZN-PT/Metglas composite. The results can be seen in Figure 5. When the dielectric constant of the PE phase is changed, the optimum PM volume fraction does not change. In the case of the magnetoelectric voltage, it is observed that when the dielectric constant is decreased by half, the magnetoelectric voltage is doubled. This indicates that the magnetoelectric voltage is inversely proportional to the dielectric constant.

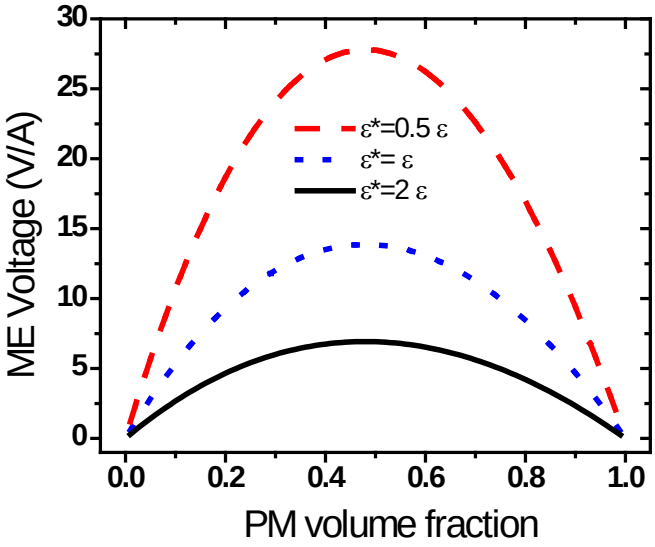

Figure 5: Magnetoelectric coupling for PZN-PT Metglas composite using different values for the PE coupling.

The influence of the elastic properties on the magnetoelectric voltage is evaluated by changing the values of the compliances of the constituents. The compliance matrix (S_{11} and S_{12}) of the PE and PM phases are set to half and twice their nominal values, to study their influence on the magnetoelectric voltage produced by the PZN-PT/Metglas composite.

The results can be seen in Figure 6. It can be seen that decreasing the values of the compliance of the PE or the PM phase results in an increase of the magnetoelectric voltage in both cases. The optimum magnetoelectric voltage is obtained at: (a) higher volume fractions when a smaller compliance of the PE phase is used and (b) lower PM volume fractions when a smaller compliance of the PM phase is used. This is due to the fact that having a smaller PM compliance requires less volume of PM phase to achieve optimum compromise between the thickness of the PE phase and the electric field generated.

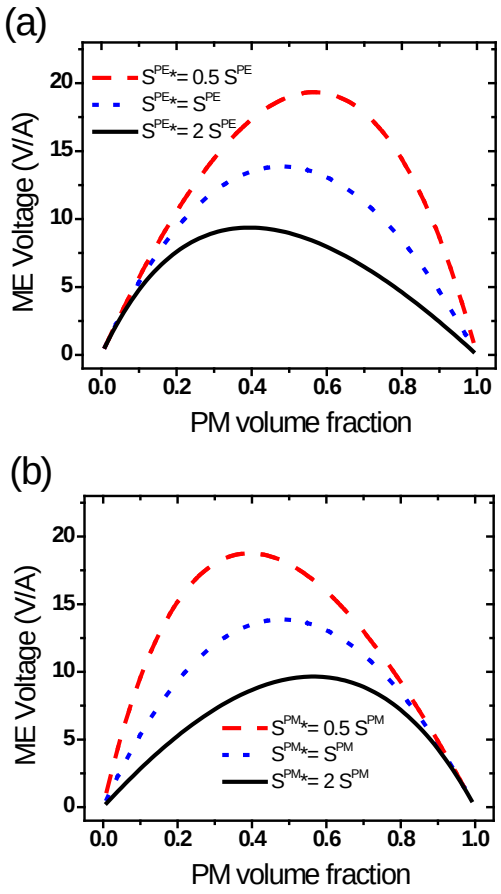

Figure 6: Magnetoelectric coupling for PZN-PT Metglas composite using different values for the mechanical compliance of: (a) PE phase and (b) PM phase.

3.4 Influence of Materials Selection

In order to compare the performance of composites made out of different combinations of PM and PE materials, the magnetoelectric voltages in TL configuration were calculated for all possible combinations between the four PE and three PM materials previously selected.

The results obtained can be seen in Figure 7. The composite structure made with Metglas and PZN-PT shows the highest magnetoelectric voltage of 13 V/A for a PM volume fraction of 47%. Even though Terfenol-D has a higher value of the PM coupling (q_{33}), which should result in a higher magnetoelectric voltage, its higher compliance results in smaller overall voltage.

The composite made with Terfenol-D and PVDF shows maximum magnetoelectric voltage of 6.9 V/A for a PM volume fraction of 25%. In this case the Terfenol-D shows a higher magnetoelectric voltage, when combined with PVDF, than Metglas. This is due to the fact that the higher PM coupling (q_{33}), has a higher benefit than the smaller compliance of Metglas.

In some applications where the weight is relevant, the use of the polymeric PVDF instead of the ceramic PZN-PT will be a better option. Additionally, PZN-PT is lead based and its use in certain countries may be restricted. Finally, PVDF is considerably less expensive than PZN-PT. Other combinations of materials possess a magnetoelectric voltage considerably smaller, without any further advantage.

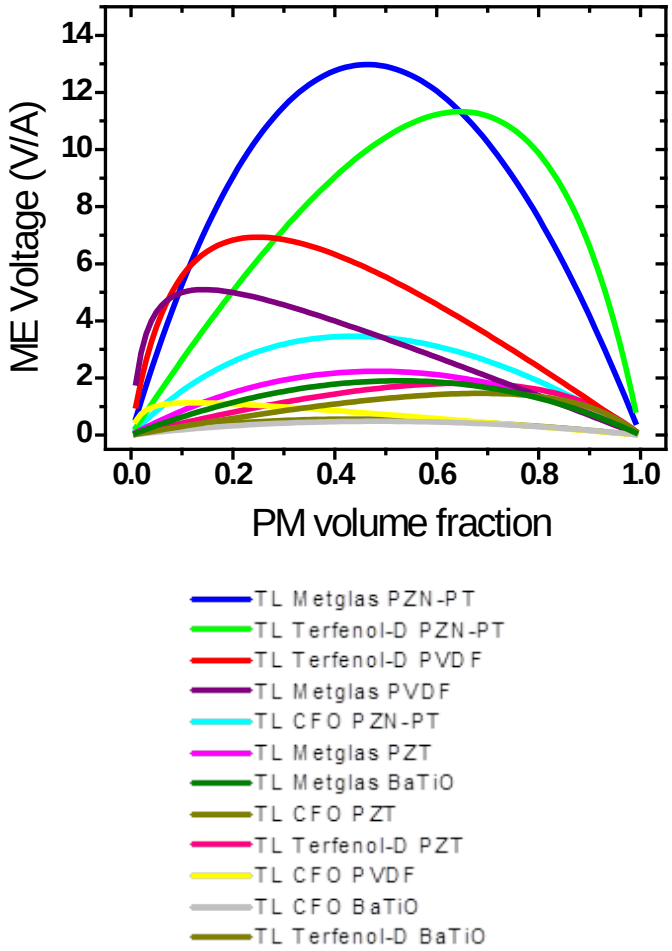

Figure 7: Magnetoelectric voltage of different composites made of the combination of the selected four PE and three PM materials for different PM volume fractions.

4. CONCLUSIONS

The optimum magnetoelectric coupling is obtained with the TL configuration when compared with TT. The magnetoelectric voltage has a linear relationship with the PE and PM coupling of the constituents and the optimum volume fraction is independent of these properties. The model shows that the magnetoelectric effect has an inverse relationship with the dielectric constant. In the case of the elastic properties, it is observed that smaller values of the compliance matrix, for the PE or PM phase, result in higher magnetoelectric voltage. Additionally, when the PM compliance is smaller than the PE compliance, the maximum magnetoelectric coupling is achieved at lower PM volume fractions. Among twelve different compositions, Metglas/PZN-PT composite with a magnetoelectric voltage of 13 V/A for a PM volume fraction of 47% and Terfenol-D/PVDF with a magnetoelectric voltage of 6.9 V/A for a PM volume fraction of 25% have the best predicted response.

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