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Effective properties via AHM-FEM for composite batteries: formulation, implementation, and validation

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The present study is part of the research project entitled Development of Autonomous Systems using multifunctional batteries for Structural Integrity Monitoring: Application in Impact Damaged Composite Structures (SAFE). The main objective is to evaluate the potential and limitations of using a new generation of multifunctional batteries in systems for monitoring the integrity of composite material structures. This project covers both micromechanical and macromechanical aspects of analyses. Micromechanical models are used to predict the effective properties of multifunctional batteries. These effective properties are then used in macromechanical models, which aim to evaluate the structural integrity of a composite structure through vibrational models and tests in conjunction with damage metrics. The main focus of the present work is the development of micromechanical models that will be used to predict the effective properties, both purely mechanical and piezo-electro-mechanical, of the materials used in multifunctional batteries. For that purpose, a periodic media is considered and it is assumed that a Representative Volume Element (RVE), or unit cell, can be used to represent the heterogeneities in the media. The Asymptotic Homogenization Method (AHM) is used as mathematical background for the derivation of the relations for the determination of the effective properties. The equilibrium relations derived from the AHM are discretized using the Finite Element Method, and all relations are implemented using in-house software. The implemented algorithms are validated by comparing them to analytical solutions and/or literature. In this way, the applicability of the micromechanical approach to determine mechanical and piezo-electro-mechanical properties, is investigated, aiming to feed the macromechanical models, which in turn will be used to evaluate the integrity of damaged composite structures. In the present stage of development of this research, the effective elastic constants of the media are presented as a function of the percentage of the PZT layer considered in the analysis.

Keywords: Asymptotic Homogenization Method, Finite Element Method, Piezoelectric Material, Effective Properties

INTRODUCTION

Structural batteries are an appealing technology used to reduce the weight, volume, and consumption of a range of vehicles and devices (Danzi et al., 2021). Those technologies integrate batteries in composite structures or use carbon fiber-based multifunctional materials. In the study of Danzi, Camanho and Braga (2021), the authors use an all-solid structural battery, which integrates an electrochemical system that can both harvest thermal and store electrical energy, while improving its mechanical performance.

Some experiments were conducted by Jacques et al. (2013) to investigate the coupling between electromechanical and mechanical properties of Lithium intercalating carbon fibers, which can be used as energy harvesting. In further investigations, Jacques et al. (2015) demonstrate that is possible to harvest electrical energy directly from mechanical work on carbon fiber bundles, by using the fibers as electrodes in a Li-ion battery cell.

The energy harvesting phenomenon can be investigated by the piezoelectric effect. The question that arises is, how to obtain the effective piezoelectric properties of a heterogeneous media, considering that the properties of the base materials are known? The effective properties of a heterogeneous media are dependent on the properties of its constituents and their volumetric fraction and spatial distribution, and micromechanical approaches are often used to tackle that kind of problem.

Among the several homogenization methods, the asymptotic homogenization method (AHM) is based on a mathematically rigorous theory and has been used in several applications in the past years. In this method, a periodic unit cell is used to represent the microstructure of the material and its effective properties are obtained through a variational formulation on a local boundary value problem (Marinelli et al., 2016). Also, as shown in previous works, the AHM can asymptotically converge to the exact or homogenized, solution by sequentially solving problems at different scales of hierarchy (Ameen, Peerlings, and Geers, 2017).

Within the Group of Aeronautical Structures (GEA - *Grupo de Estruturas Aeronautical*), a great effort has been made in the past few years to tackle several kinds of problems involving the determination of effective mechanical and piezoelectric properties of heterogeneous media, using micromechanics and multiscale methods. Here, one can emphasize the use of the Asymptotic Homogenization Method (AHM) to derive the analytical solutions to predict the effective behavior of heterogeneous materials and the use of numerical models based on a Representative Volume Element (RVE), implemented alongside the Finite Element package ABAQUS, to estimate the average behavior of the media. Some contributions of the Group of Aeronautical Structures to the topic of effective properties of piezoelectric materials are shown next.

The works of Moreno, Tita, and Marques (2009) and Moreno, Tita and Marques (2010) address a procedure based on the modeling of a Represent Volume Element (RVE) by the finite element method. The RVE is analyzed under several loading and boundary conditions to evaluate the homogenized elastic, dielectric, and piezoelectric coefficients. A discussion regarding the influence of boundary conditions on the effective material properties is also addressed. Medeiros et al. (2012) use the methodology to evaluate a globally homogeneous medium equivalent to the original composite, using a representative volume element. The method consists of the development of unit cell numerical models for smart composite materials with piezoelectric fibers made of PZT embedded in a non-piezoelectric matrix. Two scenarios are considered. In the first one, the unit cell is applied to predict the effective material coefficients of a transversely isotropic piezoelectric composite with circular cross-section fibers, with different arrangements, as shown in Fig. Medeiros et al. (2012). The second scenario consists in applying the method to calculate the equivalent properties for smart composite materials with square cross-section fibers with different arrangements, as shown in Fig. 2.

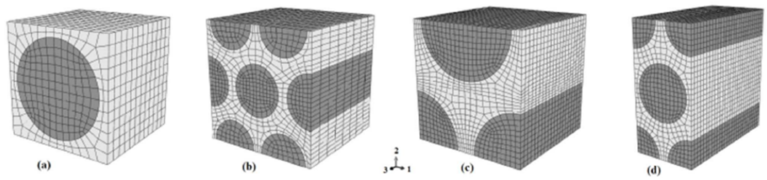

Figure 1 – Finite Element Models for circular section fiber with different arrangements: (a) square; (b) hexagonal (1); (c) hexagonal (2) and (d) hexagonal (3)

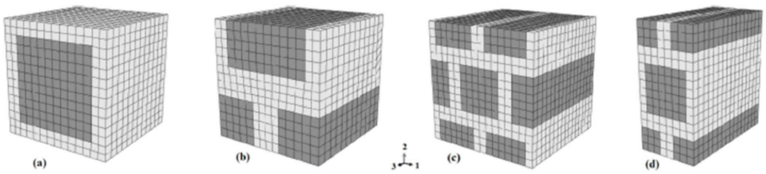

Figure 2 – Finite Element Models for square section fiber: (a) square; (b) hexagonal (1); (c) hexagonal (2) and (d) hexagonal (3)

The authors use suitable boundary conditions allowing the simulation of all modes of the overall deformation arising from any arbitrary combination of mechanical and electrical loading of the analyses performed in the RVE. The homogenized coefficients are obtained from the Theorem of Average. The results are compared to analytical solutions of the Asymptotic Homogenization Method. Although adequate results are obtained, differences up to 23% can be observed by comparing the analytical solution and the numerical solution proposed by the authors. Thus, this approach requires particular care with RVE boundary conditions.

The same approach is used by Tita et al. (2015), in which a numerical approach is used to evaluate the effective properties of piezoelectric fibers embedded in a non-piezoelectric matrix, now extending the method to consider imperfect contact between the fiber and the matrix. Transversely, isotropic piezoelectric materials with circular and square cross-section fibers are analyzed for square arrangements with different fiber volume fractions as well as with perfect and imperfect contact conditions. The RVE model used for the circular arrangement is shown in Fig. 3.

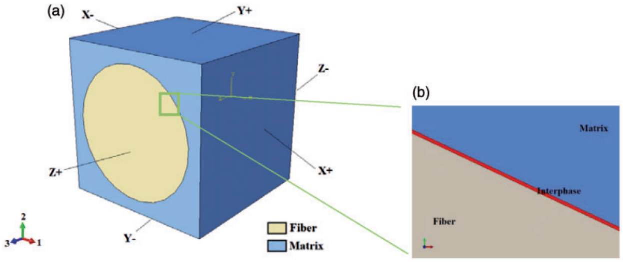

Figure 3 – Unit cell for a circular fiber arrangement and the three considered phases in detail

The results for circular and square cross-section fibers with perfect contact are compared to the literature data to verify the consistency of the proposed numerical approach.

In Brito-Santana, et al. (2016), the aforementioned approach is extended to evaluate the effective properties in piezoelectric composite materials with imperfect fiber-matrix adhesion, considering different interface models. A new imperfect interface model for a thin elastic interface is derived and the calculation of all coefficients of the material tensor and the calculation was performed via the Finite Element package ABAQUS^{TM}. A systematic procedure, written in Python language, was developed to calculate all RVE effective coefficients and the results are compared to classical interface models found in the literature. Several fiber volume fractions and interface penalization parameters are used and compared to models found in the literature.

In the work of López-Realpozo et al. (2020), the two scales asymptotic homogenization method (AHM) is applied for determining the effective coefficients of laminated piezoelectric composite with a periodic structure under non-uniform electrical and mechanical imperfect contact conditions. The analytical expressions of the local problems and the effective coefficients as a result of the AHM are explicitly derived and the results are compared with limit cases, where perfect and uniform imperfect contact conditions are considered. The model considered for the laminate, as well as the periodic cell and a cross-section of the periodic cell, are shown in Fig. 4. As the analytical solution is derived to estimate the effective properties of laminated piezoelectric composites, the model can be applied to a variety of situations concerning the quality of the interfaces between the constituents.

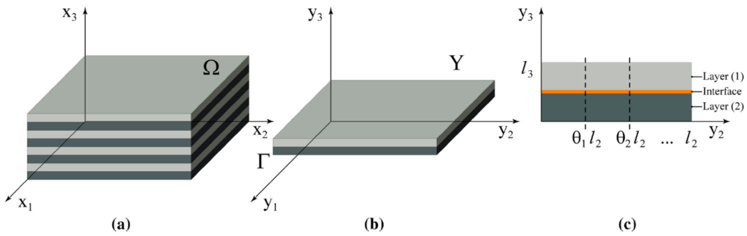

Figure 4 – (a) Laminate composite; (b) Periodic cell; (c) Partition of the interface

The influence of the piezoelectric layer position on the shear effective coefficients of the constitutive tensor of smart layered composites, subjected to delamination, is addressed in Silva et al. (2021). The model consists of a nine-layer unit cell in the mesoscale and the delamination is represented by degrading the properties of the interface between specific layers. In a macro-scale analysis, the authors show that the piezoelectric layer position has little influence over the homogenized shear coefficients.

Considering the aspects pointed out above, the objective of this work lies in the determination of the effective constitutive relations of a linear piezoelectric media. The Asymptotic Homogenization Method is considered for deriving the relations for the effective properties of a heterogeneous media, which are solved by using the Finite Element Method. The method is then used to obtain the effective piezoelectric properties of the batteries, which can later be used to feed a macroscopic model.

Thus, the outline of the present work is as follows: initially, the basic concepts concerning piezoelectric materials are presented, including the definition of the problem, equilibrium, and constitutive relations. Then, the main considerations of the Asymptotic Homogenization Method are presented, as well as the relations for the effective properties of the media. Finally, the method is applied to obtain the effective properties of a solid-state battery that has a piezoelectric behavior.

In the present stage of development of this research, the Asymptotic Homogenization Method, solved by the Finite Element Method, is used to obtain the effective elastic coefficients of the media. It is considered a three-layered Representative Volume Element, in which the intermediate layer is comprised of a PZT material. The effective properties are obtained as a function of the thickness of the PZT layer.

FORMULATION

In this section, the basic equilibrium and constitutive relations of piezoelectric materials are presented. In addition, the main considerations for the use of the Asymptotic Homogenization Method and the basic formulation of the relations of the effective properties are presented. The formulation and notation follow the works of Berger et al. (2003), Castillero et al. (1998), Rodrıguez-Ramos et al. (2001) and Bravo-Castillero et al. (2001).

Let Ω be a domain in R^3 with boundary $\Gamma = \partial\Omega$. The strain tensor, ε_{ij} and the electric field vector E_i are represented, respectively, as

$$\varepsilon_{ij} = \frac{1}{2}\left(u_{i,j} + u_{j,i}\right) \tag{1}$$

and

$$E_i = -\phi_{,i}, \tag{2}$$

where u_i is the displacement vector and ϕ is the electric potential gradient.

It is considered that the coupled electromechanical behavior of the piezoelectric media can be modeled by using a linear constitutive equation. Therefore, the linear constitutive equations for the piezoelectric materials are written as

$$\sigma_{ij} = C_{ijkl}\varepsilon_{kl} - e_{kij}E_k \tag{3}$$

$$D_i = e_{ikl}\varepsilon_{kl} + \kappa_{ik}E_k \tag{4}$$

where C_{ijkl} is the fourth-order elasticity tensor (measured in a constant electric field), e_{kij} is the piezoelectric tensor (measured at a constant strain or electric field), that relates the stress to the electric field, κ_{ik} is the dielectric tensor (measured at a constant strain), σ_{ij} is the second-order stress tensor, ε_{ij} is the second-order strain tensor, D_i is the vector of electric displacement and E_k is the electric field.

On Eq.3, the first term on the right-hand side represents the elasticity portion, while the second one is the Conv. piezoelectric term. On Eq. 4, the first term on the right-hand side represents the direct piezoelectric relation, while the second one is the permittivity.

In addition, the tensors, C_{ijkl}, e_{kij} and κ_{ij} have the classical properties of symmetry and positivity given by (Castillero et al., 1998)

$$C_{ijkl} = C_{jikl} = C_{klij}, \tag{5}$$

$$e_{kij} = e_{kji}, \tag{6}$$

$$\kappa_{ij} = \kappa_{ji} \tag{7}$$

$$\exists\eta > 0 \forall\varepsilon \in E_s^3, C_{ijkl}(x)\varepsilon_{ij}\varepsilon_{kl} \geq \eta|\varepsilon|^2 \tag{8}$$

$$\exists\eta_1 > 0 \forall a \in R^3, \varepsilon_{ij}(x)a_i a_j \geq \eta_1|a|^2 \tag{9}$$

where E_s^3 is the space of symmetric third order tensors. Considering

$$\sigma_{ij,i} = 0, \quad \sigma_{ij} = \sigma_{ji}, \quad D_{i,i} = 0, \tag{10}$$

and using Eqs. 1, 2 and 10 in Eqs. 3 and 4, yields

$$\left(C_{ijkl}u_{k,l} + e_{mij}\phi_{,m}\right)_{,j} = 0 \tag{11}$$

$$\left(e_{iml}u_{m,l} - \kappa_{im}\phi_{,m}\right)_{,i} = 0, \tag{12}$$

which represent a set of equation to obtain u_i and ϕ. Equations 11 and 12 are not sufficient, thus, the set of boundary conditions is assigned:

$$u_i = \bar{u}_i \quad on \quad \Gamma_1, \quad \sigma_{ij} n_j = t_i \quad on \quad \Gamma_2, \tag{13}$$

$$\phi_i = \bar{\phi}_i \quad on \quad \Gamma_3, \quad D_i n_i = -\bar{Q} \quad on \quad \Gamma_4 \tag{14}$$

$$\Gamma = \Gamma_1 \cup \Gamma_2, \quad \Gamma_1 \cap \Gamma_2 = \emptyset, \tag{15}$$

$$\Gamma = \Gamma_3 \cup \Gamma_4, \quad \Gamma_3 \cap \Gamma_4 = \emptyset. \tag{16}$$

Equations 11-12 subjected to the boundary conditions Eqs. 13-16 are a closed system of the static piezoelectric problem.

Now, consider a piezoelectric material with a periodic, or quasi-periodic structure. The properties of that class of material can be determined by the Asymptotic Homogenization Method. The basic of AHM, as used in this study, is to obtain the macroscopic, or average, properties of a heterogeneous media formed by a periodic, or quasi-periodic, microstructure, as a function of the microscale. In other words, by analyzing a Representative Volume Element (RVE), which is the smallest portion of the media where one can find a pattern, one can predict its effective behavior. The method consists in solving a set of equilibrium problems subjected to suitable boundary conditions to obtain the displacement field and electric potential gradient on the microscopic level, which can be used to obtain the homogenized properties of the media.

Consider Y as the size of the periodic cell, with $Y = (0, Y_1) \times (0, Y_2) \times (0, Y_3)$. In addition, consider that the material properties, C_{ijkl}, e_{mij} and κ_{im} to be Y-periodic. Here, it is assumed that that two scales describe the problem: y is the local coordinate, or fast coordinate and x is the global, or slow, coordinate, with

$$\alpha = \frac{x}{y}, \tag{17}$$

$$\alpha = \frac{l}{L} \tag{18}$$

where α is a small parameter, l is the characteristic length of the cell and L is the characteristic length of the domain.

For a Y-periodic material, the properties are functions of the fast variable, it is thus assumed that $C_{ijkl} = C_{ijkl}(y)$, $e_{kij} = e_{kij}(y)$ and $\kappa_{ij} = \kappa_{ij}(y)$. The solution of Eqs. 11-12 and Eqs. 13-16 for this kind of problem takes the form of a two-scale asymptotic expansion. Thus, the displacement field and the electric potential gradient are written as

$$u_i(x,y) = u_i^0(x,y) + \alpha u_i^1(x,y) + \alpha^2 u_i^2(x,y) + \ldots, \tag{19}$$

$$\phi_i(x,y) = \phi_i^0(x,y) + \alpha \phi_i^1(x,y) + \alpha^2 \phi_i^2(x,y) + \ldots. \tag{20}$$

The mathematical formulation leads to a set of relations for the homogenized properties of the media, given by

$$C^H_{ijpq} = \left\langle C_{ijpq}(y) + C_{ijkl}(y)_{pq} M_{k,l}(y) + e_{kij}(y)_{pq} N_{,k}(y) \right\rangle \tag{21}$$

$$e^H_{ipq} = \left\langle e_{ipq}(y) + e_{ikl}(y)_{pq} M_{k,l}(y) - \kappa_{ik}(y)_{pq} N_{,k}(y) \right\rangle \tag{22}$$

$$\kappa^H_{ip} = \left\langle \kappa_{ip}(y) - e_{ikl}(y)_p Q_{,k}(y) + \kappa_{ik}(y)_p P_{k,l}(y) \right\rangle \tag{23}$$

where ${}_{pq}M$, ${}_{pq}N$, ${}_{p}P$ and ${}_{p}Q$ are the so called local-functions, which are local auxiliary unique Y-periodic functions, independent of x (Castillero et al., 1998), the operator $\langle \quad \rangle$ is

$$\langle F \rangle = \frac{1}{|Y|} \int_Y F dY, \tag{24}$$

and the material coefficients $C_{ijpq}(y)$, $e_{ipq}(y)$ and $\kappa_{ik}(y)$ are known functions of the constituents.

This work follows the solution for the local functions, as well as the finite element discretization, reported in Berger et al. (2003).

RESULTS

This section presents the results of the research. A solid-state battery-like battery media is considered. The model is comprised of three layers in which the outer layers play the role of the electrodes and the inner layer represents the electrolyte. The determination of the effective properties of the media is performed in a finite element in-house software written in Julia Language (Bezanson et al., 2017). In this stage of the research, the focus is on the homogenized elastic properties of the media. Thus, the implemented AHM-FEM algorithm is used to predict the influence of the thickness of the PZT layer on the effective properties of a layered battery-like model.

For an initial analysis, the inner layer is considered as a PZT layer, with properties given in Tab. 1 (Berger et al., 2005). The materials considered for the electrode are metallic alloys. The mechanical properties of the Copper (Husson et al., 2008) and Zinc (Ledbetter, 1977) layers are given in Tab. 2.

Table 1 – Electroelastic properties of the PZT material.

Parameters	C_{1111}	C_{1122}	C_{1133}	C_{3333}	C_{1313}	C_{1212}	e_{311}	e_{333}	e_{223}	κ_{11}	κ_{33}
Dimension	GPa	GPa	GPa	GPa	GPa	GPa	C/m^2	C/m^2	C/m^2	pF/m	pF/m
PZT	121	75.4	75.2	111	21.1	22.8	-5.4	15.8	12.3	8.11	7.35

Table 2 – Mechanical properties of the constituents.

Parameters	E	ν
Dimension	GPa	-
Copper	120.0	0.34
Zinc	153.0	0.15

Figure 5 shows the finite element model used for the simulations, in which the middle layer represents the PZT material while the outer layers represent the Copper and Zinc materials. It is considered that the model is laminated in the y_3 direction. It is also considered a perfect bond between adjacent layers.

In the first moment, it is investigated how the mechanical properties of the model vary in terms of the volumetric fraction of the PZT material. The dimensions of the domain are considered unitary and volumetric fractions of the PZT material of 0.2, 0.4, 0.6, and 0.8 are considered. In all cases, a finite element mesh is discretized by 1000 solid isoparametric hexahedra with quadratic interpolation. No influence of the finite element mesh was observed by using those parameters.

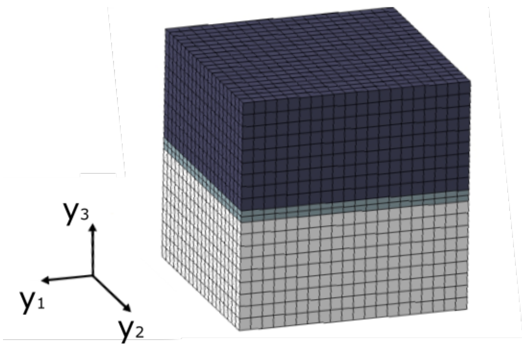

Figure 5 – Finite element model: Mesh of the RVE depicting the different layers.

Figure 6 shows the variation of the components C_{1111}, C_{3333}, C_{1122}, C_{1133}, C_{1212} and C_{1313} of the effective fourth order elasticity tensor as a function of the volumetric fraction of the PZT layer. The variation of the properties shown in Fig. 6 shows a consistency with previous investigations by the authors (see Brito-Santana et al. (2018), Brito-Santana, et al. (2019), Christoff et al. (2020) and Christoff, Brito-Santana, and Tita (2021)), which deal with the variation of the effective mechanical properties of layered materials.

It is known that the AHM is a powerful tool for mechanical analyses. The effective properties are crucial when a macromechanical model is considered. In the context of this research, the effective properties are used to feed a model to obtain the frequency response of a composite structure. The battery-like models act like a piezoelectric sensor on those structures. By considering both intact and damaged structures, one can compare the obtained frequency response, and for further analysis, estimate the severity of the damage in the structure. Thus, a reliable way to predict the effective properties of the media is important, since it is used as a sensor in the macromechanical model.

Additionally, the batteries considered in this project are manually manufactured. Thus, it is expected a deviation in the geometrical parameters of the medium, as well as a variation in the mechanical properties of the constituents. Consequently, the analysis carried out serves to predict the effective properties of the medium, even when geometric variations are considered.

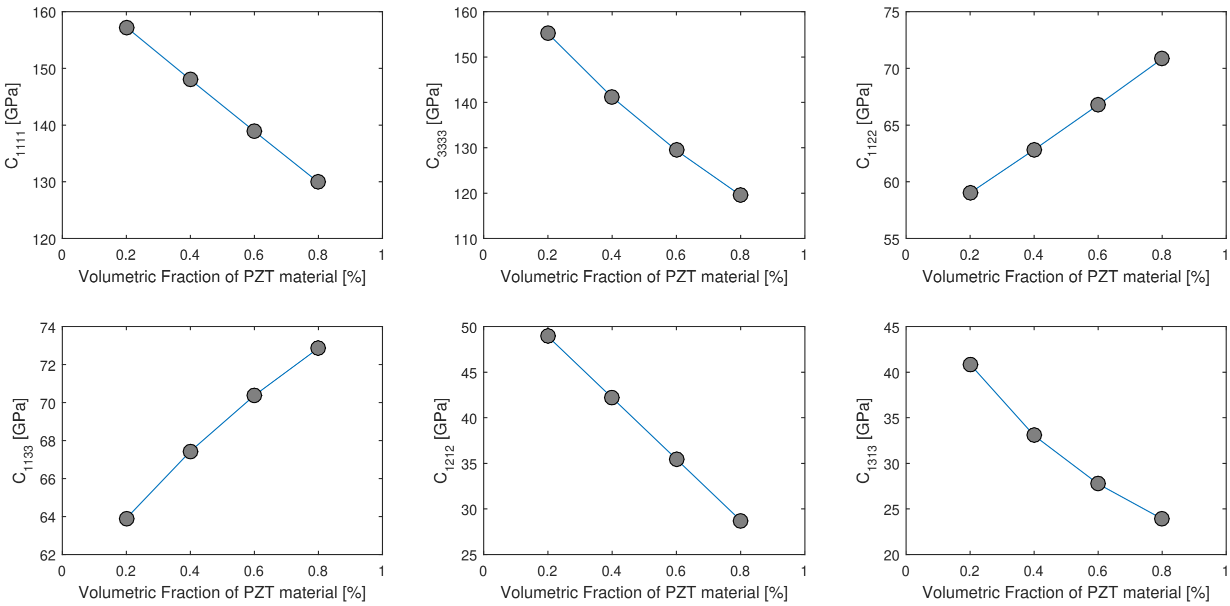

Figure 6 – Predicted elastic components as a function of the % of PZT material.

For future analyses, the AHM-FEM is considered to obtain the effective piezoelectric tensor and the dielectric tensor of the media. In addition, the influence of the geometrical parameters of the piezoelectric layer on the effective piezoelectric and dielectric properties will be investigated.

Furthermore, a sensitivity analysis can be performed to verify mathematically how the geometric parameters and the properties of the constituents influence the effective properties of the media. In a subsequent analysis, the sensitivity analysis can be extrapolated to verify the influence on the frequency response of the composite structure considered in this research project.

The final goal is to be able of predicting the frequency response of the composite structures considering the deviations in the micromechanical models.

CONCLUSIONS

The present work addresses the micromechanical aspects of piezoelectric materials. A solid-state battery with piezoelectric behavior is considered, and the determination of the effective properties of the media is investigated.

The basic relations for a piezoelectric material are presented, in which a linear constitutive equation is considered. The domain is considered heterogeneous and the effective properties of the media are obtained by the Asymptotic Homogenization Method. The finite element method is used to discretize the local functions and to estimate the effective properties of the media.

The model considered for an initial analysis is a three-layer media, in which the outer layers represent metallic electrodes and the inner layer, which represents the electrolyte, is here considered as a PZT layer. The initial results investigate the variation of the homogenized elastic constants of the media as a function of the volume fraction of the PZT layer. The results show consistency with the results presented in the literature.

For further analyses, the complete homogenized properties of the piezoelectric media can be obtained. Thus, the homogenized elastic constants, the piezoelectric constants, and the dielectric constants are investigated. In addition, sensitivity analyses can be carried out to study the influence of the micromechanical model on the effective properties of the media, and on the frequency response of a composite structure that uses the micromechanical model as a piezoelectric sensor.

The final goal is to be able of predicting the frequency response of the composite structures considering the deviations in the micromechanical models.

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