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A NEW VARIATIONAL FRAMEWORK FOR LARGE STRAIN PIEZOELECTRIC HYPERELASTIC MATERIALS

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Abstract. In this paper, a novel nonlinear variational formulation is presented for the numerical modelling of piezo-hyperelastic materials. Following energy principles, a new family of anisotropic extended internal energy density functionals is introduced, dependent upon the deformation gradient tensor and the Lagrangian electric displacement field vector. The requirement to obtain solutions to well defined boundary value problems leads to the definition of energy density functionals borrowing concepts from polyconvex elasticity. Material characterisation of the constitutive models is then carried out by means of experimental matching in the linearised regime (i.e. small strains and small electric field). The resulting variational formulation is discretised in space with the help of the Finite Element Method, where the resulting system of nonlinear algebraic equations is solved via the Newton-Raphson method after consistent linearisation. Finally, a series of numerical examples are presented in order to assess the capabilities of the new formulation.

1 INTRODUCTION

The earliest piezoelectric materials to be discovered, i.e. crystals, have shown limited applicability due to their high stiffness and brittleness. The recent advent of piezoelectric polymers has meant a turning point in the development of piezoelectricity. The circumvention of the drawbacks associated with their crystal predecessors has broadened considerably their applications as actuators, power generators and energy harvesters.

Piezoelectric polymers have traditionally been used as smart actuators in microelectromechanical systems. However, their ability to emulate the functioning of biological muscles as well as their large strain capabilities, have recently triggered the emergence of new exciting applications, such as artificial muscles. A more recent application within the field of smart actuators can be found in space microwave antennas. These devices experience shape deviations due to pre-stress and thermal expansion. These deviations are

corrected by controlling an applied electric potential on a piezoelectric patch to maintain the desired shape of the antenna reflector. Figure 1 illustrates two application examples.

Figure 1: (a) Electroactive actuated robotic arm. (b) Shape control of space antenna.

The large strain and piezoelectric capabilities associated to piezoelectric polymers confer them with attractive properties in the field of power generation and energy harvesting. There is currently a growing need for this kind of applications. For instance, piezoelectric eels (see Figure 2(a)), which are used as part of submarine devices in long endurance military missions. The energy-harvesting eel is designed to extract energy from the wake of a bluff body in an ocean current. The basic configuration is a leading bluff body trailed by a thin flexible piezoelectric eel. The bluff body generates vortices which excite a flapping motion of the eel. The eel deformation results in strain of the piezoelectric membrane, which in turn generates a voltage across the material. Other current applications include small piezoelectric devices that when attached to the insole of a shoe are able to transform mechanical motion into electrical power (see Figure 2(b)).

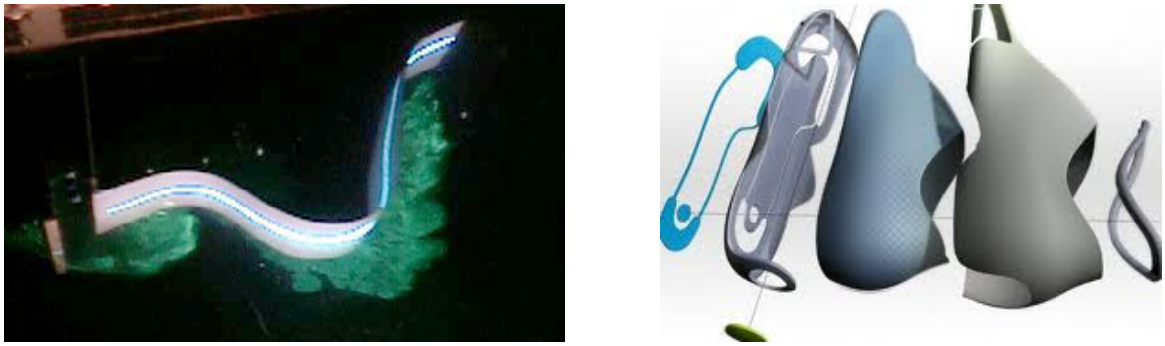


Figure 2: (a) Piezoelectric eel. (b) Walking energy harvesting device.

Electromechanical interactions in materials are not only due to piezoelectricity. Another important phenomenon which has found a vast number of applications within the field of electromechanical actuators is electrostriction. This phenomenon results from a quadratic relation between stresses and electric field. The application of electric fields on purely electrostrictive materials would lead to a deformation on the material. However, unlike piezoelectricity, this effect cannot be reversed. It is worth emphasising that piezoelectricity and electrostriction are not mutually exclusive.

The existing framework for the numerical simulation of piezoelectric materials requires an enhancement as a result of the development of these new polymers, capable of undergoing large deformations. Unlike crystals, the classical linearised theory can no longer be applied for a reliable computer simulation. In this paper, a nonlinear variational formulation for piezo-hyperelastic materials is introduced with the help of the internal energy density U constructed on the basis of the right Cauchy-Green deformation tensor $\boldsymbol{C}$ and the Lagrangian electric displacement field vector $\boldsymbol{D}_0$.

2 ANISOTROPIC STRUCTURE OF PIEZOELECTRIC MATERIALS.

The proposed internal energy density U must lead to the definition of a constitutive model which needs to capture the material response in an accurate manner. Accordingly, U must be compliant with the physical and mathematical constraints inherent to these materials (e.g. anisotropy). This will be taken into consideration by means of a hybrid approach which combines the isotropic extension surface concept introduced in reference [2] and a more recent methodology proposed in reference [3].

The first approach, i.e. the isotropic extension concept, introduces the so-called isotropic extension surface ϕ, which includes the set of vectors and tensor valued functions which are preserved under the action of the material symmetry group which characterises the corresponding anisotropy of the material, that is,

$$\phi = \phi\left(\boldsymbol{C}, \boldsymbol{D}_0\right). \tag{1}$$

As a result, the creation of anisotropic invariants can alternatively be achieved by formulating isotropic invariants which take into consideration the isotropic extension surface ϕ. Thus, the extended internal energy density functional U is created in terms of isotropic invariants of the following arguments,

$$U = \hat{U}\left(\boldsymbol{C}, \boldsymbol{D}_0, \phi\left(\boldsymbol{C}, \boldsymbol{D}_0\right)\right). \tag{2}$$

An alternative approach for the creation of anisotropic invariants was recently proposed in [3]. In this reference, a new metric tensor $\boldsymbol{G} = \boldsymbol{H}\boldsymbol{H}^{\mathrm{T}}$ is introduced, where $\boldsymbol{H}$ represents a linear tangent map established between the Cartesian base triad $\{\boldsymbol{e}_1, \boldsymbol{e}_2, \boldsymbol{e}_3\}$ and the crystallographic base system $\{\boldsymbol{a}_1, \boldsymbol{a}_2, \boldsymbol{a}_3\}$, the latter related to the associated Bravais lattice (see Figure 3).

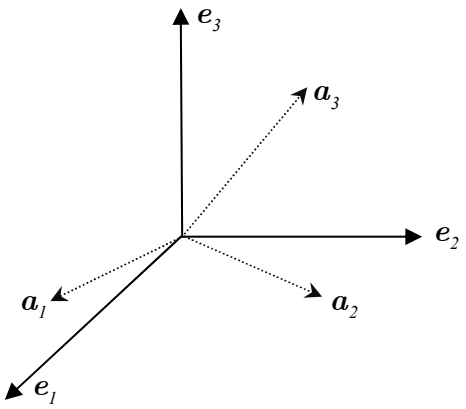


Figure 3: Crystallographic $\{\boldsymbol{a}_1, \boldsymbol{a}_2, \boldsymbol{a}_3\}$ and Cartesian $\{\boldsymbol{e}_1, \boldsymbol{e}_2, \boldsymbol{e}_3\}$ base vectors.

3 CONSTITUTIVE EQUATIONS IN ELECTRO-MECHANICS.

The derivation of the constitutive equations for Electro-Mechanics requires a revision of the First and Second laws of Thermodynamics. Following an integral material description, the First law of Thermodynamics gives the variation of internal energy as [1],

$$\begin{aligned} \frac{d}{dt}\int_{\Omega_0} U\left(\boldsymbol{F}, \boldsymbol{D}_0, \eta_0\right)\, d\Omega_0 &= \int_{\Omega_0} \boldsymbol{P} : \dot{\boldsymbol{F}}\, d\Omega_0 + \int_{\Omega_0} \boldsymbol{E}_0 \cdot \dot{\boldsymbol{D}}_0\, d\Omega_0 \\ &- \int_{\partial\Omega_0} \boldsymbol{\nabla}_0 \cdot \boldsymbol{q}_0\, d\Omega_0 + \int_{\Omega_0} R_0\, d\Omega_0, \end{aligned} \tag{3}$$

where $\boldsymbol{F}$ is the deformation gradient tensor, $\boldsymbol{D}_0$ is the Lagrangian electric displacement field, η_0 is the Lagrangian entropy, $\boldsymbol{P}$ is the first Piola-Kirchoff stress tensor, $\boldsymbol{E}_0$ is the Lagrangian electric field, θ is the temperature, $\boldsymbol{q}_0$ is the Lagrangian Fourier-Stokes heat flux vector field and R_0 is the Lagrangian heat supply field per unit volume.

The Clausius-Duhem form of the Second law of Thermo-Electro-Mechanics can be written as [1],

$$\frac{d}{dt}\int_{\Omega_0} \eta_0\, d\Omega_0 \geq \int_{\Omega_0} \frac{R_0}{\theta}\, d\Omega_0 - \int_{\Omega_0} \boldsymbol{\nabla}_0 \cdot \frac{\boldsymbol{q}_0}{\theta}\, d\Omega_0. \tag{4}$$

Combination of both First and Second laws leads to,

$$\begin{aligned} &\int_{\Omega_0} \left(\boldsymbol{P} - \frac{\partial U}{\partial \boldsymbol{F}}\right) : \dot{\boldsymbol{F}}\, d\Omega_0 + \int_{\Omega_0} \left(\boldsymbol{E}_0 - \frac{\partial U}{\partial \boldsymbol{D}_0}\right) \cdot \dot{\boldsymbol{D}}_0\, d\Omega_0 \\ &+ \int_{\Omega_0} \left(\theta - \frac{\partial U}{\partial \eta_0}\right) \dot{\eta_0}\, d\Omega_0 - \int_{\Omega_0} \frac{1}{\theta} \boldsymbol{\nabla}_0 \theta \cdot \boldsymbol{q}_0\, d\Omega_0 \geq 0. \end{aligned} \tag{5}$$

Constitutive equations based on the internal energy U can be derived from the combination of the First and Second law of Thermo-Electro-Dynamics [1],

$$\boldsymbol{P} = \frac{\partial U\left(\boldsymbol{F}, \boldsymbol{D}_0, \eta_0\right)}{\partial \boldsymbol{F}} \quad \boldsymbol{E}_0 = \frac{\partial U\left(\boldsymbol{F}, \boldsymbol{D}_0, \eta_0\right)}{\partial \boldsymbol{D}_0} \quad \theta = \frac{\partial U\left(\boldsymbol{F}, \boldsymbol{D}_0, \eta_0\right)}{\partial \eta_0} \quad -\frac{1}{\theta}\boldsymbol{\nabla}_0\theta \cdot \boldsymbol{q}_0 \geq 0, \tag{6}$$

where $\boldsymbol{q}_0$ can be related to the material gradient of the temperature θ as,

$$\boldsymbol{q}_0 = -\boldsymbol{k}\boldsymbol{\nabla}_0\theta, \tag{7}$$

where $\boldsymbol{k}$ is the thermal conductivity tensor. Positive definiteness of $\boldsymbol{k}$ ensures that $-\frac{1}{\theta}\boldsymbol{\nabla}_0\theta \cdot \boldsymbol{q}_0 \geq 0$. For reversible Electro-Mechanics, the constitutive equations can be simplified as,

$$\boldsymbol{P} = \frac{\partial U\left(\boldsymbol{F}, \boldsymbol{D}_0\right)}{\partial \boldsymbol{F}} \quad \boldsymbol{E}_0 = \frac{\partial U\left(\boldsymbol{F}, \boldsymbol{D}_0\right)}{\partial \boldsymbol{D}_0}. \tag{8}$$

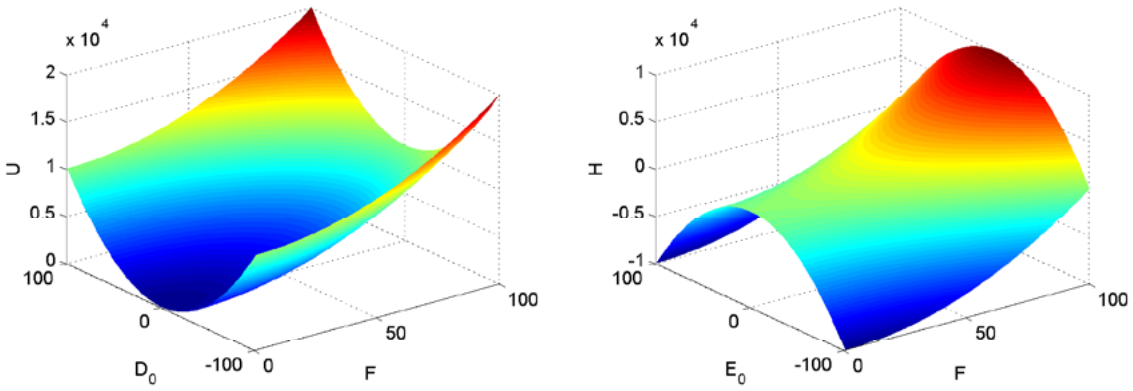
Different constitutive laws could be proposed by introducing alternative internal energy densities U. In addition, the electric enthalpy $H\left(\boldsymbol{F}, \boldsymbol{E}_0\right)$ can be introduced by means of the Legendre transform,

$$H\left(\boldsymbol{F}, \boldsymbol{E}_0\right) = U\left(\boldsymbol{F}, \boldsymbol{D}_0\right) - \boldsymbol{E}_0 \cdot \boldsymbol{D}_0. \tag{9}$$

For the electric enthalpy, analogous constitutive laws to equations (8) can be written as,

$$\boldsymbol{P} = \frac{\partial H\left(\boldsymbol{F}, \boldsymbol{E}_0\right)}{\partial \boldsymbol{F}} \quad \boldsymbol{D}_0 = -\frac{\partial H\left(\boldsymbol{F}, \boldsymbol{E}_0\right)}{\partial \boldsymbol{E}_0}. \tag{10}$$

In the abscence of external electric or mechanical loads, equilibrium configurations are reached when the internal energy $U\left(\boldsymbol{F}, \boldsymbol{D}_0\right)$ attains a minimum. It is possible to show that for the same equilibrium configuration, the complementary electric enthalpy $H\left(\boldsymbol{F}, \boldsymbol{E}_0\right)$ attains a saddle point. In other words, in a neighbourhood of the equilibrium configuration, the internal energy $U\left(\boldsymbol{F}, \boldsymbol{D}_0\right)$ would be a convex function of $\boldsymbol{F}$ and $\boldsymbol{D}_0$ whereas the electric enthalpy $H\left(\boldsymbol{F}, \boldsymbol{E}_0\right)$ would be a saddle function, i.e convex in $\boldsymbol{F}$ and concave in $\boldsymbol{D}_0$. Figure 3 shows the nature of both $U\left(\boldsymbol{F}, \boldsymbol{D}_0\right)$ and $H\left(\boldsymbol{F}, \boldsymbol{E}_0\right)$ in a neighbourhood of the equilibrium configuration for a one-dimensional case.



(a) Internal energy (b) Electric enthalpy

Figure 4: Behaviour of different types of energy densities in the vicinity of an equilibrium configuration.

4 Mathematical requirements: Polyconvexity.

The properties of the constitutive models used for the numerical simulation of piezoelectric polymers must also guarantee existence of solutions to boundary value problems in large deformation regimes. Convexity of the internal energy functional ensures existence. However, this condition is too stringent as it precludes the appearance of different equilibrium configurations with equal potential energy, i.e. buckling. Moreover, it can also violate material frame indifference. A necessary and almost sufficient condition for the existence of minimisers to a variational formulation is quasiconvexity. However, this is an integral condition whose verification is a cumbersome task. As a result, the conditions preferred in this work which guarantee the existence of solutions to well defined boundary value problems, are polyconvexity and coercivity [4]. Borrowing concepts from nonlinear elasticity, an energy density functional is said to be polyconvex in the deformation gradient tensor $\boldsymbol{F}$ if it can be written as a convex function of the following arguments,

$$U = \hat{U}\left(\boldsymbol{F}, \operatorname{Cof}\boldsymbol{F}, \det\boldsymbol{F}\right). \tag{11}$$

An intuitive and physical understanding of polyconvexity can be gained from its geometric interpretation. The arguments appearing in equation (11) govern the transformations by means of which line, area and volume material elements are mapped to their spatial counterparts (see Figure 5).

There is an extensive work done in the creation of polyconvex energy functionals in the field of nonlinear elasticity. However, the coupled nature of the electromechanical problem requires a generalisation of this concept. As a result, this paper presents a methodology for the development/implementation of polyconvex energy functionals on the basis of the deformation gradient tensor $\boldsymbol{F}$ and the material electric displacement field $\boldsymbol{D}_0$ [5]. According to this methodology, polyconvexity is enforced by means of a convex function

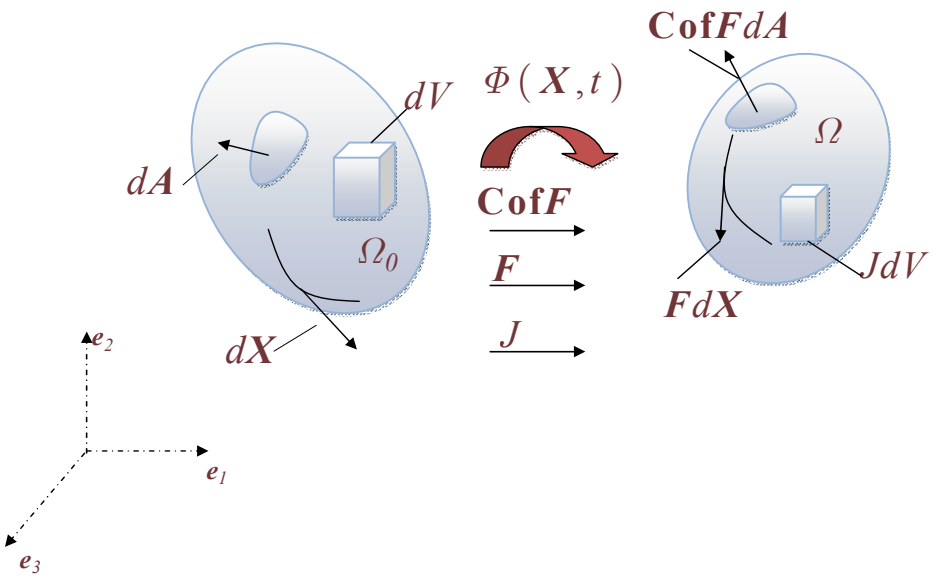


Figure 5: Deformation mapping and relevant kinematic operators ($\boldsymbol{F}, \mathrm{Cof}\,\boldsymbol{F}, J = \det \boldsymbol{F}$).

of the following arguments,

$$U = \hat{U}\left(\boldsymbol{F}, \mathrm{Cof}\boldsymbol{F}, \det\boldsymbol{F}, \boldsymbol{D}_0\right). \tag{12}$$

5 Constitutive models for piezo-hyperelastic materials.

From the physical point of view, a possible decomposition of the internal energy density is carried out in terms of the three constitutive parts involved in the formulation, i.e. mechanical, piezoelectric and electrostrictive,

$$U\left(\boldsymbol{D}_0, \boldsymbol{C}\right) = U_m\left(\boldsymbol{C}\right) + U_e\left(\boldsymbol{D}_0, \boldsymbol{C}\right) + U_p\left(\boldsymbol{D}_0, \boldsymbol{C}\right), \tag{13}$$

where U_m, U_p and U_e account for the mechanical, piezoelectric and electrostrictive components of the stored internal energy functional U, respectively. Naturally, the mechanical component of the energy functional can also be split into its isotropic and anisotropic contributions,

$$U_m = U_{iso,m}\left(\boldsymbol{C}\right) + U_{aniso,m}\left(\boldsymbol{C}\right). \tag{14}$$

The isotropic contribution of the mechanical component $U_{iso,m}\left(\boldsymbol{C}\right)$ can be chosen from any of the existing polyconvex strain energy functionals, i.e. Mooney-Rivlin. The definition of the three remaining components (see equations (13)-(14)) completes the characterisation of the material model. Anisotropic mechanical energy functionals built on the basis of polyconvex invariants can be found in [3]. This author proposes the follow-

ing expression for $U_{aniso,m}$ in terms of an additive decomposition of a series of families j characterising the anisotropy of the material,

$$U_{aniso,m} = \sum_j \mu_j \left(\frac{{J_{4_j}}^{\alpha_j+1}}{\alpha_j+1} + \frac{J_{5_j}^{\beta_j+1}}{\beta_j+1} + \frac{tr\left(\boldsymbol{G}_j^m\right) III_{\boldsymbol{C}}^{-\gamma_j}}{\gamma_j} \right), \tag{15}$$

with,

$$J_{4_j} = \text{tr}\left(\boldsymbol{C}\boldsymbol{G}_j^m\right); \quad J_{5_j} = \text{tr}\left(\boldsymbol{C}\,\text{Cof}\,\boldsymbol{G}_j^m\right); \quad III_{\boldsymbol{C}} = \det\boldsymbol{C}, \tag{16}$$

where $\boldsymbol{G}_j^m$ is a possible metric tensor characterising the anisotropy of the material, α_j, β_j and γ_j are dimensionless parameters and μ_j is a mechanical material parameter. For the material model to be complete, it is necessary to account for the remaining constitutive counterparts, i.e. piezoelectric and electrostrictive.

In this paper, a novel definition for the respective components of the energy functional on the basis of polyconvex invariants is introduced. A possible electrostrictive component U_e would be,

$$U_e = \sum_j \omega_j \left[\boldsymbol{p}_{e,j} \cdot \boldsymbol{C}\boldsymbol{p}_{e,j} + I_{\boldsymbol{C}}^2 + \left(\boldsymbol{p}_{e,j} \cdot \boldsymbol{p}_{e,j}\right)^2 \right] + f_e\left(\boldsymbol{C}\right) + g_e\left(\boldsymbol{\mathcal{D}}_0\right), \tag{17}$$

with,

$$\boldsymbol{p}_{e,j} = m_j \boldsymbol{G}_j^e \boldsymbol{\mathcal{D}}_0 \quad \boldsymbol{\mathcal{D}}_0 = \frac{\boldsymbol{D}_0}{\sqrt{\mu_{ref}\varepsilon_0}}, \tag{18}$$

where $\boldsymbol{G}_j^e$ is a possible metric tensor characterising the anisotropy of the material, m_j is a dimensionless parameter, ω_j is an electrotrictive material parameter, ε_0 is the electric permittivity in vacuum and μ_{ref} is a reference mechanical material parameter. A possible contribution for the piezoelectric component U_p would be,

$$U_p = \sum_{j,k} \lambda_{jk} \left[\boldsymbol{p}_{p,jk} \cdot \boldsymbol{C}\boldsymbol{p}_{p,jk} + I_{\boldsymbol{C}}^2 + \left(\boldsymbol{p}_{p,jk} \cdot \boldsymbol{p}_{p,jk}\right)^2 \right] + f_p\left(\boldsymbol{C}\right) + g_p\left(\boldsymbol{\mathcal{D}}_0\right), \tag{19}$$

with,

$$\boldsymbol{p}_{p,jk} = s_j \boldsymbol{G}_j^p \boldsymbol{\mathcal{D}}_0 + \boldsymbol{N}_k, \tag{20}$$

where $\boldsymbol{G}_j^p$ is a possible metric tensor characterising the anisotropy of the material, s_j is a dimensionless parameter, $\boldsymbol{N}_k$ accounts for the structural vectors included in the isotropic extension surface of the material considered and λ_{jk} is a piezoelectric material parameter. It is important to emphasise that the definitions of the mechanical U_m and electrostrictive U_e components are based upon the methodology proposed in [3]. However, the piezoelectric component has been formulated by means of a combination of this method and the isotropic extension concept in [2].

The functions f_e, f_p, g_e and g_p are chosen so that no stress or electric field is obtained in the origin,

$$\begin{aligned}\boldsymbol{S}|_{\boldsymbol{D}_0=\boldsymbol{0},\boldsymbol{C}=\boldsymbol{I}} &= 2\frac{\partial U_p}{\partial \boldsymbol{C}}\bigg|_{\boldsymbol{D}_0=\boldsymbol{0},\boldsymbol{C}=\boldsymbol{I}} + 2\frac{\partial U_e}{\partial \boldsymbol{C}}\bigg|_{\boldsymbol{D}_0=\boldsymbol{0},\boldsymbol{C}=\boldsymbol{I}} = \boldsymbol{0}\\ \boldsymbol{E}_0|_{\boldsymbol{D}_0=\boldsymbol{0},\boldsymbol{C}=\boldsymbol{I}} &= \frac{\partial U_p}{\partial \boldsymbol{D}_0}\bigg|_{\boldsymbol{D}_0=\boldsymbol{0},\boldsymbol{C}=\boldsymbol{I}} + \frac{\partial U_e}{\partial \boldsymbol{D}_0}\bigg|_{\boldsymbol{D}_0=\boldsymbol{0},\boldsymbol{C}=\boldsymbol{I}} = \boldsymbol{0}.\end{aligned} \tag{21}$$

The complete characterisation of the material model defined through the internal energy density requires the definition of the material properties α_j, β_j, γ_j, μ_j, ω_j, m_j, λ_{jk}, s_j and the metric tensors $\boldsymbol{G}_j^m$, $\boldsymbol{G}_j^e$ and $\boldsymbol{G}_j^p$. This is achieved by performing a match in the origin, i.e. $\boldsymbol{C}=\boldsymbol{I}, \boldsymbol{E_0}=\boldsymbol{0}$, between the constitutive tensors derived from the proposed energy functionals and the experimental tensors available in the linearised regime (i.e. small strains and small electric field),

$$\boldsymbol{\mathcal{C}}|_{\boldsymbol{C}=\boldsymbol{I},\boldsymbol{E}_0=\boldsymbol{0}} = \boldsymbol{c}; \quad \boldsymbol{\mathcal{P}}|_{\boldsymbol{C}=\boldsymbol{I},\boldsymbol{E}_0=\boldsymbol{0}} = \boldsymbol{p}; \quad \boldsymbol{\mathcal{A}}|_{\boldsymbol{C}=\boldsymbol{I},\boldsymbol{E}_0=\boldsymbol{0}} = \boldsymbol{\epsilon}, \tag{22}$$

where $\boldsymbol{\mathcal{C}}$, $\boldsymbol{\mathcal{P}}$ and $\boldsymbol{\mathcal{A}}$ are the elastic, piezoelectric, and dielectric material tensors, respectively, derived from the energy functional as follows,

$$\begin{aligned}\mathcal{A}_{ij}\left(\boldsymbol{C},\boldsymbol{E}_0\right) &= \left[\frac{\partial^2 U\left(\boldsymbol{C},\boldsymbol{D}_0\right)}{\partial \boldsymbol{D}_0 \partial \boldsymbol{D}_0}\right]^{-1}_{ij}\\ \mathcal{P}_{ijk}\left(\boldsymbol{C},\boldsymbol{E}_0\right) &= -2\mathcal{A}_{mi}\frac{\partial^2 U\left(\boldsymbol{C},\boldsymbol{D}_0\right)}{\partial C_{jk}\partial D_{0_m}}\\ \mathcal{C}_{ijkl}\left(\boldsymbol{C},\boldsymbol{E}_0\right) &= 4\frac{\partial^2 U\left(\boldsymbol{C},\boldsymbol{D}_0\right)}{\partial C_{ij}\partial C_{kl}} + 2\frac{\partial^2 U\left(\boldsymbol{C},\boldsymbol{D}_0\right)}{\partial C_{ij}\partial D_{0_m}}\mathcal{P}_{mkl}.\end{aligned} \tag{23}$$

From the previous match, the necessary material parameters can be obtained either by identification or minimisation.

6 Numerical results.

The resulting variational formulation is discretised in space with the help of the Finite Element Method, where the resulting system of nonlinear algebraic equations is solved

via the Newton-Raphson method after consistent linearisation. In this section, a series of numerical examples are presented in order to demonstrate the robustness and applicability of the formulation.

Figure 6 shows an example in which the application of an external electric field produces a deformation in a composite shell. Figure 7 depicts another example of actuator application. The application of an external electric field on a composite material produces deformations and changes in shape. In this case, the different anisotropic orientation of both layers of materials leads to an out of plane deformation. This effect enables these materials to be designed to carry out a specific mechanically demanded task.

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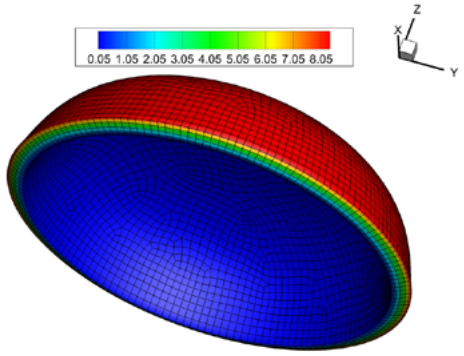
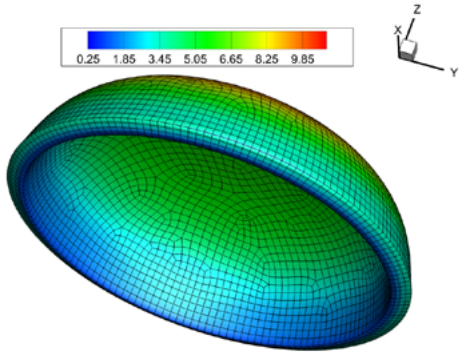
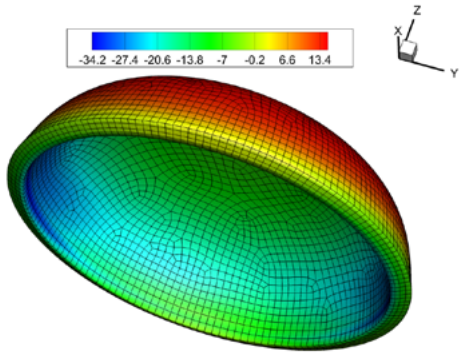


(a) Electric potential (V)

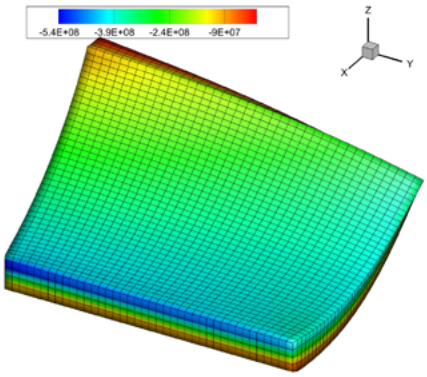


(b) Electric field in OZ direction (V/m)

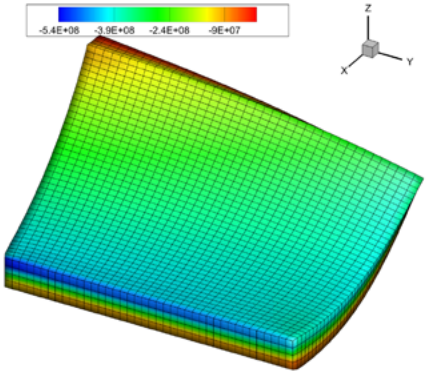


(c) Stress $\boldsymbol{\sigma_{xx}}$ (N/m^2)

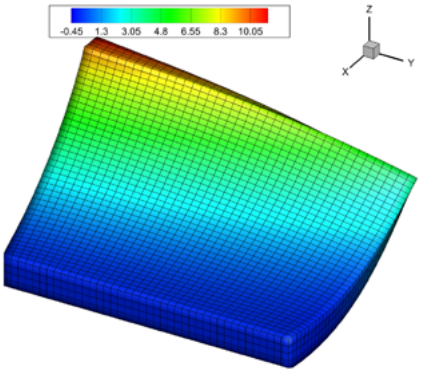
Figure 6: Composite circular shell subject to electric potential gradient across the thickness.



(a) Electric potential (V)



(b) Electric displacement in OX direction (N/mV)



(c) Displacement in OZ direction (m)

Figure 7: Composite rectangular shell subject to electric potential gradient across the thickness.