Modelling dielectric composites in finite deformation elasticity

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Keywords: Dielectric elastomers, Composite materials, Soft actuators.

SUMMARY. We present some preliminary results regarding the response of a two-phase, rank-1 laminated dielectric composite subjected to large deformations, where the stress state in each phase is obtained in terms of macroscopic deformation gradient and macroscopic nominal stress. It is shown that, for some lamination angles, the performance of the composite –in terms of actuation strain– is improved with respect to the homogeneous case.

1 INTRODUCTION

The class of electroactive polymers (EAPs) provides attractive advantages: they are soft, lightweight, undergo large deformations, possess fast response time and are resilient. However, widespread application has been hindered by their limitations: the need for large electric field, relatively small forces and energy density. It is now recognized that the limitations arise from poor electromechanical coupling in typical polymers. This in turn is related to the fact that the typical polymers have a small ratio of dielectric to elastic modulus. Recent experimental findings [1, 2] suggest that these difficulties can be resolved with the aid of composites made out of flexible matrices with inclusions of high dielectric materials.

Analysis of the governing equations for the electromechanical response of composites undergoing finite deformations was initiated in [3]. In this work, starting from the fundamental balance laws for the energy stored in the composite, and assuming that the energy-density functions for the individual phases are known, we derive expressions for the stress state in each phase in terms of macroscopic deformation gradient and macroscopic nominal stress. In a way of an example, we determine actuation strains [4] of a laminated composite, demonstrating that indeed these strains can be improved by considering composite dielectrics. We compare our findings with the corresponding results of [3] that were determined by direct solution of the governing equations for the same class of composites.

The theory of deformable dielectrics has been firmly established starting from the contributions by Toupin [5] and Tiersten [6] and, more recently, by other authors (see, *e.g.*, [7, 8, 9]). Even though different formulations can be presented, it is based on the notion of a free-energy density H, sometimes called 'electric enthalpy', where the independent entries are the deformation gradient $\boldsymbol{F}$ and the nominal electric field $\boldsymbol{E}^\circ$, so that the constitutive equations for an incompressible material can be formulated as (see the Appendix for a synopsis of the governing equations of a composite dielectric under finite strain and for the notation used in this paper)

$$\boldsymbol{S} = \frac{\partial H}{\partial \boldsymbol{F}} - p\boldsymbol{F}^{-T}, \quad \boldsymbol{D}^\circ = -\frac{\partial H}{\partial \boldsymbol{E}^\circ}, \tag{1}$$

where $\boldsymbol{S}$ is the First Piola-Kirchhoff (or nominal) stress tensor and $\boldsymbol{D}^{\circ}$ is the nominal electric displacement vector.

2 HOMOGENEOUS MATERIAL

In this section we carry out the analytic solution of the coupled B.V.P. associated with an electroactive actuator. We consider a strip of a homogeneous dielectric material, with an unstressed thickness h°, lying between two parallel and flexible electrodes with fixed potentials ϕ^{+} and ϕ^{-}. To this end we make the following assumptions:

1. The two electrodes remain straight and parallel during the deformation of the actuator.
2. The electrodes are flexible with a negligible elastic moduli and thus do not extract mechanical traction on the dielectric layer.
3. We consider the deformation of the actuator due to electromechanical coupling but with no external loads. Accordingly, the traction boundary condition is $\boldsymbol{t} = \boldsymbol{0}$.
4. The size of the circumferential boundaries of the layer is considerably smaller than the size of the top and bottom boundaries which are in contact with the electrodes. Thus, we neglect edge or fringing effects due to the potential field induced by the electrodes outside the actuator, and assume that the electric field outside the dielectric vanishes identically.

The electric field in the reference configuration (*i.e.*, $\boldsymbol{E}^{\circ} = -\mathsf{Grad}\,\phi$) due to the potential difference is

$$\boldsymbol{E}^{\circ} = [0, E_2^{\circ}]\,, \tag{2}$$

where E_2° is constant.

In a way of an example, we use the following expression for the energy-density function of the electro-active polymer

$$H(\boldsymbol{F}, \boldsymbol{E}^{\circ}) = \frac{\mu}{2}(I_1 - 3) - \frac{\varepsilon_0 \varepsilon}{2} I_{5e}, \tag{3}$$

where the invariants appearing in (3) are defined as $I_1 = \mathsf{tr}\boldsymbol{C}$ and $I_{5e} = \boldsymbol{E}^{\circ} \cdot (\boldsymbol{C}^{-1}\boldsymbol{E}^{\circ})$.

Consider the boundary condition (38), in the case with no mechanical traction and neglecting fringing field effects (*i.e.*, $\boldsymbol{\sigma}_0$), we get $\boldsymbol{\sigma}\hat{\boldsymbol{n}} = \boldsymbol{0}$, for all $\hat{\boldsymbol{n}}$, therefore $\boldsymbol{\sigma} = \boldsymbol{0}$. From the constitutive equations, the total stress associated with (3) is

$$\boldsymbol{\sigma} = \mu \boldsymbol{F}\boldsymbol{F}^T + \varepsilon_0 \varepsilon \boldsymbol{E} \otimes \boldsymbol{E} - p\boldsymbol{I} = \boldsymbol{0}, \tag{4}$$

and the electric displacement is

$$\boldsymbol{D} = \varepsilon_0 \varepsilon \boldsymbol{E}. \tag{5}$$

Assuming a deformation gradient of the form

$$\boldsymbol{F} = \begin{pmatrix} F_{11} & F_{12} \\ 0 & \frac{1}{F_{11}} \end{pmatrix}, \tag{6}$$

we obtain the solution

$$\begin{aligned} F_{12} =& 0, \\ F_{11} =& \left[1 - \frac{\varepsilon_0 \varepsilon}{\mu}\left(\frac{\Delta\phi}{h^{\circ}}\right)^2\right]^{-\frac{1}{4}}. \end{aligned} \tag{7}$$

As the electric field squeezes the strip along direction x_2, the component F_{11} represents the stretch along the actuation direction x_1 ($F_{11} > 1$).

3 A RANK-1 LAMINATED COMPOSITE

We consider now a heterogeneous dielectric electroactive actuator, with the same dimensions as in the previous examples. In addition to the hypotheses presented in Sect. 2, we add the following assumptions:

1. The characteristic size of the heterogeneity is much smaller than the size of the actuator.
2. The morphology of the actuator is such that the heterogeneous dielectric is macroscopically homogeneous.

We note that with the above assumptions the boundary conditions applied to the actuator are such that if it was made out of a homogeneous material the electrical fields within the actuator were uniform. These type of boundary conditions are commonly being used to determine the effective properties of composite materials. It can be shown that if the potential difference between the two electrodes is $\hat{\phi} = -\boldsymbol{E}_0 \cdot \boldsymbol{x}$, then the mean electric field

$$\boldsymbol{E} \equiv \frac{1}{V} \sum_{r=1}^{n} \int_{B^{(r)}} \boldsymbol{E}^{(r)} dV = \boldsymbol{E}_0, \tag{8}$$

where V is the volume of the composite in the deformed configuration. Since we assumed that the composite is macroscopically homogeneous, to determine the electric fields developing in the composite it is sufficient to consider a unit volume element (in the deformed configuration) which is representative of the composite microstructure and yet considerably smaller than the overall size of the actuator. We require that within the unit volume element $\boldsymbol{E} = \boldsymbol{E}_0$, and thus ensure that the far-field boundary condition is satisfied in an average sense. With this requirement we need to solve Maxwell's equations (27) and (28) in the unit element together with the continuity conditions (29), (30) and (31) and the constitutive relation (1) for a given composite. A parallel procedure can be applied to obtain the macroscopic deformation gradient $\boldsymbol{F}$.

We consider a rank-1 composite made out of two incompressible neo-Hookean phases in volume fractions $\lambda^{(1)}$ and $\lambda^{(2)} = 1 - \lambda^{(1)}$, respectively. The two phases are characterized by elastic shear moduli $\mu^{(1)}$ and $\mu^{(2)}$, and dielectric constants $\varepsilon^{(1)}$ and $\varepsilon^{(2)}$. The lamination direction is singled out by the unit vector normal to the layers plane $\hat{\boldsymbol{n}}$ (in the current state). The electrical field continuity equation along the interface is

$$\boldsymbol{E}^{(1)} \hat{\boldsymbol{m}} = \boldsymbol{E}^{(2)} \hat{\boldsymbol{m}}, \tag{9}$$

where $\boldsymbol{E}^{(i)}$ is the electric field in phase (i) of the material and $\hat{\boldsymbol{m}}$ is an arbitrary unit vector in the layers' plane (both are in the current state). This equation can be expressed alternatively as

$$[\![\boldsymbol{E}]\!] = \boldsymbol{E}^{(2)} - \boldsymbol{E}^{(1)} = k\hat{\boldsymbol{n}}, \tag{10}$$

where k is a scalar. The average electric field in the material (again, in the current state) can be defined as

$$\boldsymbol{E} = \lambda^{(1)} \boldsymbol{E}^{(1)} + \lambda^{(2)} \boldsymbol{E}^{(2)}. \tag{11}$$

Since the quantities we know by measuring or activating is the overall deformation gradient $\boldsymbol{F}$ and the applied electric field $\boldsymbol{E}$, we need to use them when expressing the nominal stress. Therefore, in this stage we *pull back* equations (10) and (11) to the reference state by using the relation $\boldsymbol{E}^\circ = \boldsymbol{F}^T \boldsymbol{E}$ to get

$$(\boldsymbol{F}^{(2)})^{-T} \boldsymbol{E}^{\circ(2)} - (\boldsymbol{F}^{(1)})^{-T} \boldsymbol{E}^{\circ(1)} = k \boldsymbol{F}^{-T} \hat{\boldsymbol{n}}^\circ, \tag{12}$$

and

$$\boldsymbol{F}^{-T} \boldsymbol{E}^\circ = \lambda^{(1)} \boldsymbol{F}^{-T} \boldsymbol{E}^{\circ(1)} + \lambda^{(2)} \boldsymbol{F}^{-T} \boldsymbol{E}^{\circ(2)}, \tag{13}$$

where $\hat{\boldsymbol{n}}^\circ$ is a unit vector normal to the phases interface pulled-back to the reference. We get from these equations

$$\begin{aligned} \boldsymbol{E}^{(1)} &= \boldsymbol{E} - \lambda^{(2)} k \boldsymbol{F}^{-T} \hat{\boldsymbol{n}}^\circ, \\ \boldsymbol{E}^{(2)} &= \boldsymbol{E} + \lambda^{(1)} k \boldsymbol{F}^{-T} \hat{\boldsymbol{n}}^\circ. \end{aligned} \tag{14}$$

In a similar approach [10] it can be shown that the relations between the phases deformation gradients and the average deformation gradient $\boldsymbol{F}$ are in the form:

$$\begin{aligned} \boldsymbol{F}^{(1)} &= \boldsymbol{F}(\boldsymbol{I} + \lambda^{(2)} \omega \hat{\boldsymbol{m}}^\circ \otimes \hat{\boldsymbol{n}}^\circ), \\ \boldsymbol{F}^{(2)} &= \boldsymbol{F}(\boldsymbol{I} - \lambda^{(1)} \omega \hat{\boldsymbol{m}}^\circ \otimes \hat{\boldsymbol{n}}^\circ). \end{aligned} \tag{15}$$

The effective strain energy is in the form

$$\bar{H}(\boldsymbol{F}, \boldsymbol{E}^\circ, \omega, k) = \lambda^{(1)} H^{(1)}(\boldsymbol{F}, \boldsymbol{E}^\circ, \omega, k) + \lambda^{(2)} H^{(2)}(\boldsymbol{F}, \boldsymbol{E}^\circ, \omega, k), \tag{16}$$

where $\omega(\boldsymbol{F})$ and $k(\boldsymbol{F}, \boldsymbol{E})$ are scalar parameters, which can be evaluated from the strain continuity and electric displacement continuity on the interface, respectively (see [3]). Alternatively, these parameters should agree with the equations [10]

$$\frac{\partial \bar{H}}{\partial \omega} = \frac{\partial \bar{H}}{\partial k} = 0. \tag{17}$$

In either way, we get for ω and k the following expressions, respectively

$$\omega = \frac{\mu^{(2)} - \mu^{(1)}}{\lambda^{(1)} \mu^{(2)} + \lambda^{(2)} \mu^{(1)}} \frac{(\boldsymbol{F} \hat{\boldsymbol{n}}^\circ) \cdot (\boldsymbol{F} \hat{\boldsymbol{m}}^\circ)}{(\boldsymbol{F} \hat{\boldsymbol{m}}^\circ) \cdot (\boldsymbol{F} \hat{\boldsymbol{m}}^\circ)}, \tag{18}$$

$$k = \frac{\varepsilon^{(1)} - \varepsilon^{(2)}}{\lambda^{(1)} \varepsilon^{(2)} + \lambda^{(2)} \varepsilon^{(1)}} \frac{\boldsymbol{E} \cdot (\boldsymbol{F}^{-T} \hat{\boldsymbol{n}}^\circ)}{(\boldsymbol{F}^{-T} \hat{\boldsymbol{n}}^\circ) \cdot (\boldsymbol{F}^{-T} \hat{\boldsymbol{n}}^\circ)}. \tag{19}$$

Let us assume an effective electric enthalpy for phase (i) in the form

$$H^{(i)}(\boldsymbol{F}, \boldsymbol{E}^\circ) = \frac{\mu^{(i)}}{2}(I_1^{(i)} - 3) - \frac{\varepsilon_0 \varepsilon^{(i)}}{2} I_{5e}^{(i)}, \tag{20}$$

where

$$I_1^{(i)} = \mathrm{Tr}[(\boldsymbol{F}^{(i)})^T \boldsymbol{F}^{(i)}] = \mathrm{Tr}[(\boldsymbol{I} \pm \lambda^{(3-i)} \omega \hat{\boldsymbol{n}}^\circ \otimes \hat{\boldsymbol{m}}^\circ) \boldsymbol{C} (\boldsymbol{I} \pm \lambda^{(3-i)} \omega \hat{\boldsymbol{m}}^\circ \otimes \hat{\boldsymbol{n}}^\circ)], \tag{21}$$

and

$$I_{5e}^{(i)} = \boldsymbol{E}^{\circ(i)} (\boldsymbol{F}^{(i)})^{-1} (\boldsymbol{F}^{(i)})^{-T} \boldsymbol{E}^{\circ(i)}. \tag{22}$$

An explicit expression for the nominal macroscopic total stress can be calculated by equation (1) to yield

$$\begin{aligned}\boldsymbol{S} =& \lambda^{(1)} \left(\mu^{(1)} \boldsymbol{F} (\boldsymbol{I} + \lambda^{(2)} \omega \hat{\boldsymbol{m}}^\circ \otimes \hat{\boldsymbol{n}}^\circ) + \boldsymbol{\sigma}^{(1)}_{MX} \boldsymbol{F}^{-T} \right) + \\ & \lambda^{(2)} \left(\mu^{(2)} \boldsymbol{F} (\boldsymbol{I} - \lambda^{(1)} \omega \hat{\boldsymbol{m}}^\circ \otimes \hat{\boldsymbol{n}}^\circ) + \boldsymbol{\sigma}^{(2)}_{MX} \boldsymbol{F}^{-T} \right) - p \boldsymbol{F}^{-T},\end{aligned} \tag{23}$$

where the expressions for the Maxwell stress $\boldsymbol{\sigma}^{(i)}_{MX}$ in the two phases are

$$\boldsymbol{\sigma}^{(1)}_{MX} = \varepsilon_0 \varepsilon^{(1)} (\boldsymbol{E} - \lambda^{(2)} k \boldsymbol{F}^{-T} \hat{\boldsymbol{n}}^\circ) \otimes (\boldsymbol{E} - \lambda^{(2)} k \boldsymbol{F}^{-T} \hat{\boldsymbol{n}}^\circ), \tag{24}$$

$$\boldsymbol{\sigma}^{(2)}_{MX} = \varepsilon_0 \varepsilon^{(2)} (\boldsymbol{E} + \lambda^{(1)} k \boldsymbol{F}^{-T} \hat{\boldsymbol{n}}^\circ) \otimes (\boldsymbol{E} + \lambda^{(1)} k \boldsymbol{F}^{-T} \hat{\boldsymbol{n}}^\circ). \tag{25}$$

This expression for the effective nominal stress tensor of the composite agrees with the results presented in [3] obtained solving directly the boundary-value problem.

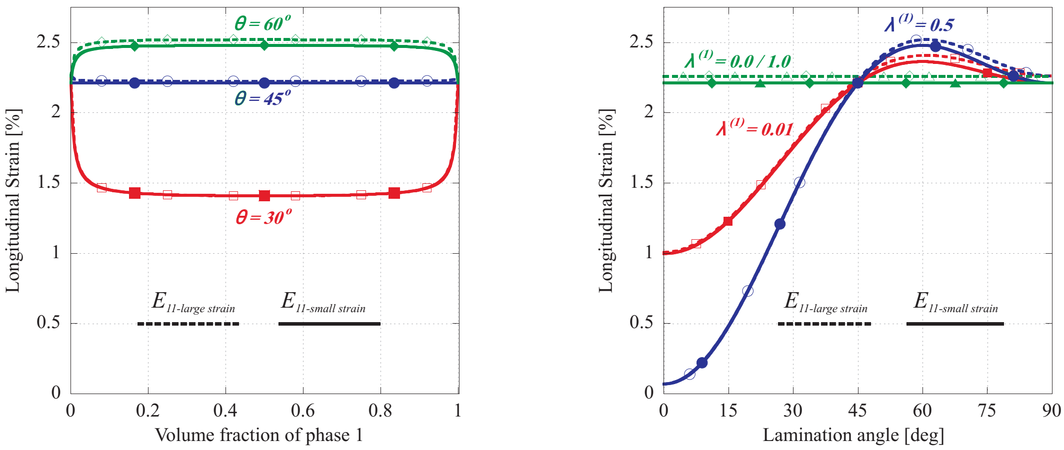

Figure 1: Overall longitudinal strain of a rank-1 laminated dielectric composite actuator (geometry described in Sect. 2) as functions of the volume fraction and lamination angle of the layers. The material parameters are $\mu^{(1)} = 1000$ MPa, $\mu^{(2)} = 8$ MPa, $\varepsilon^{(1)} = 1000$, $\varepsilon^{(2)} = 8$ and the applied electric field is $E = 100$ MV/m.

The longitudinal (actuation) strain (along direction x_1) of the heterogeneous actuator is reported in Fig. 1 in terms of volume fraction of phases (left) and lamination angle (right). The parameters of the two dielectrics (shear modulus and dielectric constants) are taken in such a way the actuation strain would be the same if the device was homogeneous, of either phase 1 or 2 (approx. 2.2 %). It is shown that while volume fraction has small influence on the overall response, the actuation strain is very sensitive on the lamination angle. For angles greater than 45° the composite gives a better performance with respect to the homogeneous case for all volume fractions.

APPENDIX: GOVERNING EQUATIONS OF A DIELECTRIC, COMPOSITE BODY

The governing equations of a dielectric composite are described in the quasi-(electro)static limit, so that no electromagnetic effects will be accounted for. $\boldsymbol{x}$ denotes points of the solid belonging to the current configuration B, that is composed of n phases ($B = \cup_{j=1}^{n} B^{(j)}$); $B^{(0)} = \mathbb{R}^3 - B$ is the external domain of the composite, therefore ∂B is the interface between B and $B^{(0)}$, while a generic interface between phases j and k ($j, k = 1, \dots, n; j < k$) will be denoted by Ξ_{jk} ($\Xi = \cup_{jk} \Xi_{jk}$). Interfaces Ξ_{jk} and $\partial B = \cup_{k=1}^{n} \Xi_{0k}$ are assumed to be sufficiently regular so that a unit normal $\boldsymbol{n}$ is defined almost everywhere pointing to j and away of k ($j < k$). Across interfaces, appropriate jump conditions must be enforced. To this end, the jump operator $[\![\,\cdot\,]\!] = (\,\cdot\,)^j - (\,\cdot\,)^k$ ($j < k$) will be adopted. The boundary ∂B can be subdivided in regions ∂B_t, where (mechanical) surface tractions $\boldsymbol{t}$ are applied, and ∂B_e, where an electrode sets a constant electric potential ϕ with a free-charge density equal to ω.

Under the action of an external electric field, a polarization field $\boldsymbol{P}(\boldsymbol{x})$ arises within B. If $\boldsymbol{E}(\boldsymbol{x})$ indicates the current electric field, $\phi(\boldsymbol{x})$ the electric potential, and

$$\boldsymbol{D}(\boldsymbol{x}) = \varepsilon_0 \boldsymbol{E}(\boldsymbol{x}) + \boldsymbol{P}(\boldsymbol{x}) \tag{26}$$

the electric displacement field, the equations of electrostatics write

$$\boldsymbol{E} = -\mathsf{grad}\,\phi, \quad \text{in } B \cup B^{(0)}, \tag{27}$$

$$\mathsf{div}\boldsymbol{D} = 0, \quad \text{in } B \cup B^{(0)}, \tag{28}$$

as free charge per unit volume vanishes, whereas jump conditions across interfaces are

$$[\![\boldsymbol{D}]\!] \cdot \boldsymbol{n} = 0, \quad \text{across } \Xi, \tag{29}$$

$$[\![\boldsymbol{D}]\!] \cdot \boldsymbol{n} = \omega, \quad \text{across } \partial B_e. \tag{30}$$

The tangential component of the electric field $\boldsymbol{E}$ is continuous across interface, therefore

$$\boldsymbol{n} \times [\![\boldsymbol{E}]\!] = \boldsymbol{0}, \quad \text{across } \Xi \cup \partial B_e, \tag{31}$$

that means

$$[\![\boldsymbol{E}]\!] = ([\![\boldsymbol{E}]\!] \cdot \boldsymbol{n})\boldsymbol{n}. \tag{32}$$

A reference configuration B°, whose points are labelled as $\boldsymbol{x}^\circ$, is also defined together with a deformation map $\boldsymbol{\chi}$ such that $\boldsymbol{x} = \boldsymbol{\chi}(\boldsymbol{x}^\circ)$ and $\boldsymbol{F} = \mathsf{Grad}\boldsymbol{\chi}$ ($B_\circ^{(0)} = \mathbb{R}^3 - B^\circ$).

In the absence of (mechanical) body forces, local equations of equilibrium can be expressed as

$$\mathsf{div}\boldsymbol{\sigma} = \boldsymbol{0}, \quad \boldsymbol{\sigma} = \boldsymbol{\sigma}^T, \quad \text{in } B, \tag{33}$$

$$\mathsf{div}\boldsymbol{\sigma}_0 = \boldsymbol{0}, \quad \boldsymbol{\sigma}_0 = \boldsymbol{\sigma}_0^T, \quad \text{in } B^{(0)}, \tag{34}$$

where $\boldsymbol{\sigma}$ is the 'total stress' in the medium[1], while the divergence-free tensor $\boldsymbol{\sigma}_0$ is the Maxwell stress in vacuum, where only electrical effects associated with $\boldsymbol{E}$ are present, of the form

$$\boldsymbol{\sigma}_0 = \varepsilon_0 \boldsymbol{E} \otimes \boldsymbol{E} - \frac{\varepsilon_0}{2}(\boldsymbol{E} \cdot \boldsymbol{E})\boldsymbol{I}. \tag{36}$$

[1] The tensor $\boldsymbol{\sigma}$ can be formally decomposed into two parts ($\boldsymbol{\sigma} = \boldsymbol{\sigma}_{MC} + \boldsymbol{\sigma}_{MX}$), where the second term, called 'Maxwell stress' by Toupin, corresponds to

$$\boldsymbol{\sigma}_{MX} = \varepsilon_0 \boldsymbol{E} \otimes \boldsymbol{E} + \boldsymbol{E} \otimes \boldsymbol{P} - \frac{\varepsilon_0}{2}(\boldsymbol{E} \cdot \boldsymbol{E})\boldsymbol{I} = \boldsymbol{E} \otimes \boldsymbol{D} - \frac{\varepsilon_0}{2}(\boldsymbol{E} \cdot \boldsymbol{E})\boldsymbol{I}, \tag{35}$$

with the property $\mathsf{div}\,\boldsymbol{\sigma}_{MX} = \mathsf{grad}\boldsymbol{E}\boldsymbol{P}$.

Across interfaces, jump conditions write

$$[\![\boldsymbol{\sigma}]\!]\boldsymbol{n} = \boldsymbol{0} \quad \text{across } \Xi \cup \partial B_e, \tag{37}$$

$$[\![\boldsymbol{\sigma}]\!]\boldsymbol{n} + \boldsymbol{t} = \boldsymbol{0} \quad \text{across } \partial B_t, \tag{38}$$

where $\boldsymbol{t}$ is the applied mechanical traction.

The Lagrangian version of the equilibrium equations follows directly from the framework described above and is based on the 'first Piola-Kirchhoff stress' $\boldsymbol{S} = J\boldsymbol{\sigma}\boldsymbol{F}^{-T}$, while Maxwell equations can be written in terms of the Lagrangian or nominal electric field and electric displacement

$$\boldsymbol{E}^{\circ} = \boldsymbol{F}^{T}\boldsymbol{E}, \quad \boldsymbol{D}^{\circ} = J\boldsymbol{F}^{-1}\boldsymbol{D}, \tag{39}$$

respectively. These fields satisfy a set of "reference electrostatic equations" (see reference [7]).

ACKNOWLEDGEMENTS. The support of GNFM-INdAM (grant "Progetto Giovani 2008") is gratefully acknowledged.

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