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### Optimisation of hierarchical dielectric elastomer laminated composites

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### ABSTRACT

This paper is concerned with the optimisation of the actuation response of electro-elastic, rank-two laminates obtained laminating a core rank-one composite with a soft phase which constitutes the shell. The analysis is performed for two classes of composites that are subjected to traction-free boundary-value problems. The results are compared with those computed in a previous study where the optimisation was carried out at small strains. The non-linear approach allows a better estimation of the geometric layout of the reference configuration to enhance the maximum stretch or shear strain at the operative applied voltage. The optimum layouts are in general characterised by a very low volume fraction of the shell while in the core the two components are almost equally distributed. The amplification of the effective local electro-mechanical response.

#### 1. Introduction

The application of electro-elasticity theory of soft dielectrics [1,2] to the area of heterogeneous materials has already shown that suitably designed microstructured composites may potentially exhibit remarkably improved actuation properties with respect to those displayed by the homogeneous materials employed so far, mainly acrylic elastomers and silicones [3–14]. Some of these studies also investigated the shortcomings associated with the use of composites that are related to peculiar phenomena such as the triggering of micro-, macro-scopic and electro-mechanical instabilities, electric field amplification inside some constituents, etc. [4,5,8–10,12].

A significant part of the research on heterogeneous soft dielectrics has been devoted to (hierarchical) layered composites. Working in the small-strain setting, Tian et al. [7] have shown that two-phase dielectric rank-*n* laminate layouts (with n > 1) can be found to improve the electromechanical actuation strain of more than one order of magnitude at the same voltage (being the gain strongly related to the order *n* of the microstructure). Or, in other words, that, in a rank-two laminate, the actuation enhancement obtained at an increasing contrast in the electromechanical properties of phases is more than proportional, almost linear in a graph where actuation strain and contrast are reported with a logarithmic scale.

In the more appropriate finite-strain, non-linear framework, laminated composites have been studied by some authors extending the methodology set out by deBotton [15]: deBotton et al. [3] provided the first preliminary analysis of the behaviour of rank-one laminates; Bertoldi and Gei [4], Rudykh and deBotton [5], Gei et al. [9], Rudykh et al. [10] and Spinelli and Lopez-Pamies [12] investigated in detail the homogenisation and the micro- and macro-scopic stability of layered composites. An evaluation of the benefit of the hierarchical structure of a rank-two composite in the electro-mechanical actuation has been carried out by Rudykh et al. [8] who have found a ten-fold improvement of the electro-mechanical coupling for a prototype laminate obtained by reinforcing with polyaniline an acrylic elastomer matrix.

This paper deals with the optimisation of the actuation response of rank-two dielectric laminates in the non-linear framework of finite electro-elasticity under both plane strain and three-dimensional unconstrained boundary conditions. The main goals are:

– to extend the analysis of the optimisation of rank-two laminates presented in [7] to better assess the gain in large-strain actuation with respect to both the homogeneous and the rank-one response;

- to establish the optimum configurations of the composite for different contrasts between the soft matrix and the stiff reinforcement;

 – to show that those configurations strongly depend on the maximum operational electric field chosen for the actuator;

– to evaluate the amplification of the current electric field in some representative configurations to provide an indication of the effective local electro-mechanical response.

We are not concerned here with occurrence of any instability or other failure mode along the electro-mechanical actuation path, but we recognise that any departure from the homogeneous response may have a strong impact in the evaluation of the optimum layout.

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#### flexible electrodes $x_1$ (co) sh a) (co) sh a) (co) sh b) (co) sh b) (co) sh a) (co) sh b) (co) sh co) s

**Fig. 1.** Geometry of the reference configuration  $B^0$  of a two-phase, ranktwo layered dielectric actuator subjected to a voltage difference  $\Delta\phi$  applied between electrodes. The close-up view represents the studied rank-two laminate highlighting the *core* rank-one structure and the *shell* composed of the soft matrix *b*. The initial thickness of the actuator is  $h^0$ . Axis  $x_3$  is the out-of-plane direction, while angles  $\theta_{co}$  and  $\theta_{sh}$  are positive counter-clockwise.

Two material combinations are considered in the examples: one is that adopted by Tian et al. [7], characterised by a matrix whose electromechanical properties match those of a polyurethane, for which the authors analysed several configurations with equal contrast between shear moduli and electric permittivities; the other possesses a softer, siliconelike matrix, reinforced with a stiffer phase which may correspond to a poly(vinylidene fluoride) electrostrictive polymer [16].

### 2. Background and homogenised response of laminated composites

The general layout of the hierarchical composite actuator under investigation is represented in Fig. 1, where  $h^0$  is the reference thickness. The laminate is constructed layering two phases: *a*, the reinforcement, and b, the matrix, usually softer than phase a and with a lower electric permittivity, in a manner consistent to the scheme called 'tree a' in [7]. According to this scheme, the final layout is obtained layering a parent rank-one composite (the core, abbreviated to 'co', whose volume fraction will be denoted by  $c^{co}$ ) with a layer of soft material acting as a *shell* (abbreviated to 'sh', with volume fraction  $c^{sh}$  such that  $c^{sh} + c^{co} = 1$ ). In [7], this layout has proven to give better performance than that of another type of arrangement, called 'tree b'. It is clear that while the materials to be mixed are two, namely, a and b, the device is composed of three parts, i.e. a,  $b^{co}$  and  $b^{sh}$ , with  $b^{sh}$  coinciding with the shell. To complete the description of the notation related of volume fractions,  $c_a^{co}$ and  $c_b^{co}$  denote, in turn, the volume fractions of a and b in the core, whilst  $c^a = c_a^{co}$  and  $c^b = c_b^{co} + c^{sh}$  are the total volume fractions of the two phases  $(c^a + c^b = 1)$ . The grades with respect to the axis  $x_1$ of the interfaces between a and b in the core and of the shell correspond to  $\theta_{co}$  and  $\theta_{sh}$ , respectively. Separation of length-scales is assumed so that homogenisation at each rank can be invoked. The theory here recalled for the solution of the electro-elastic problem for hierarchical composites relies on the formulation that can be found in [6,11,12,15], for which the effective properties can be obtained exactly up to a set of algebraic equations.

A notation which is standard in large-strain electro-elasticity is adopted.<sup>1</sup> Symbols  $D^0$  and  $E^0$  denote, in general, nominal electric displacement and nominal electric fields, respectively, while *S* and *F* 

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stand for the total first Piola–Kirchhoff stress tensor and the deformation gradient, respectively. As they were introduced, without any subscript, those four quantities are referred to the *macroscopic* response of the composite whereas, when accompanied by a subscript, they are referred to the indicated constituent so, e.g.  $F_k = \text{Grad } \mathbf{x}_k$  ( $k = a, b^{\text{co}}, b^{\text{sh}}$ ) denotes the local deformation gradient that, together with the local nominal electric field  $E_k^0$ , is assumed to be constant in each part. Both phases *a* and *b* obey an extended neo-Hookean strain-energy function formulated for incompressible materials, namely

$$W_k = \frac{\mu_k}{2} (F_k \cdot F_k - 3) - \frac{\epsilon_k}{2} (F_k^{-T} E_k^0 \cdot F_k^{-T} E_k^0) \quad (k = a, b^{\text{co}}, b^{\text{sh}}),$$
(1)

where  $\mu_k$ ,  $\varepsilon_k$  represent the local shear modulus and electric permittivity, respectively. Clearly,  $W_{b^{\text{sh}}} = W_{b^{\text{co}}} = W_b$ ,  $\mu_{b^{\text{sh}}} = \mu_{b^{\text{co}}} = \mu_b$  and  $\varepsilon_{b^{\text{sh}}} = \varepsilon_{b^{\text{co}}} = \varepsilon_b$ . In each part, the electro-elastic constitutive equations (see [1]) provide the local response

$$S_k = \frac{\partial W_k}{\partial F_k} - p_k F_k^{-T}, \quad D_k^0 = -\frac{\partial W_k}{\partial E_k^0} \quad (k = a, b^{\text{co}}, b^{\text{sh}}),$$
(2)

where  $p_k$  is the arbitrary constant linked to the incompressibility constraint. Similarly to  $F_k$  and  $E_k^0$ ,  $S_k$ ,  $D_k^0$  and  $p_k$  are constant in each part, as well.

The voltage  $\Delta\phi$  applied between the compliant electrodes will induce a macroscopic transverse electric field directed along  $x_2$  whose nominal value  $E^0$  corresponds to  $\Delta\phi/h^0$ , i.e.  $E^0 = \Delta\phi/h^0 e_2$ , where  $e_2$  is the unit vector directed along axis  $x_2$ . The actuation response of the device can be achieved formally by solving two rank-one problems, that within the core and the macroscopic one involving core and shell. However, it is not necessary to solve in full the two tasks as we can exploit a remarkable result obtained by Spinelli and Lopez-Pamies [12] where an electro-elastic strain-energy function was directly formulated for a rankone laminate, whose phases follow Eq. (1), in terms of a set of invariants which takes into account the electro-mechanical anisotropic response of the material. This energy, here denoted by  $W_{\rm co}$ , has an expression reported in the Appendix .

Based on this observation, the macro-scale, rank-one problem involving core and shell (Fig. 1) can be solved by exploiting the following jumps at the interface whose normal pointing towards the shell is  $n^0$ , namely,

$$(F_{co} - F_{sh})m^{0} = \mathbf{0}, \quad (S_{co} - S_{sh})n^{0} = \mathbf{0},$$
$$(D_{co}^{0} - D_{ch}^{0}) \cdot n^{0} = 0, \quad n^{0} \times (E_{co}^{0} - E_{ch}^{0}) = \mathbf{0},$$
(3)

and the following constitutive equations, similar to (2),

$$S_i = \frac{\partial W_i}{\partial F_i} - p_i F_i^{-T}, \quad D_i^0 = -\frac{\partial W_i}{\partial E_i^0} \quad (i = \text{co, sh}),$$

where the notation is now obvious. It is only worth noting now that parameter  $p_{co}$  can be set as  $p_{co} = p_a c^a + p_{b^{co}} c_b^{co}$ .

The relationships between local and macroscopic responses can be obtained by noting that, on the one hand, the underlying theory of composites at finite strain demands

$$F = c^{\rm co} F_{\rm co} + c^{\rm sh} F_{\rm sh}, \qquad E^0 = c^{\rm co} E^0_{\rm co} + c^{\rm sh} E^0_{\rm sh}; \tag{4}$$

on the other, interface compatibility  $(\mathbf{3})_1$  must be satisfied while  $(\mathbf{3})_4$  requires

$$E_{\rm co}^0 - E_{\rm sh}^0 = \beta n^0.$$
 (5)

As a consequence, admissible representations of  $F_i$ ,  $E_i^0$  (i = co, sh) turn out to be

$$F_{\rm co} = F\left(I + c^{\rm sh}\alpha \otimes n^0\right), \quad F_{\rm sh} = F\left(I - c^{\rm co}\alpha \otimes n^0\right)$$
(6)

and

$$E_{\rm co}^0 = E^0 + c^{\rm sh} \beta n^0, \quad E_{\rm sh}^0 = E^0 - c^{\rm co} \beta n^0, \tag{7}$$

<sup>&</sup>lt;sup>1</sup> see [9]. Boldface letters  $x^0$  and x represent points of  $B^0$  and of the current configuration B, respectively, while differential operators with capital letters are evaluated with reference to  $B^0$ .

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