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Enhanced soft dielectric composite generators: the role of ceramic fillers

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Abstract

A notable issue in the field of dielectric elastomers is the characterization of composite materials with improved electromechanical coupling destined for mechanical-to-electrostatic energy converters. To this aim, random composites, where ceramic fillers with high dielectric constant are dispersed in a silicone matrix, represent an interesting option. Currently, the most promising reinforcing materials to be immersed in a silicone matrix, already tested for soft dielectric actuators, are lead magnesium niobate–lead titanate (PMN–PT) and lead zirconate-titanate (PZT). To estimate the performance improvement entailed by the composite device with respect to the homogeneous matrix, a typical four-phase cycle is considered in the model, where nominal load and electric charge are alternately held constant. Different materials are being studied: a composite consisting of a matrix in poly-dimethyl-siloxane (PDMS) reinforced with PMN–PT, assuming a filler concentration of 10% in volume and a PDMS–PZT composite with a 1% volume fraction of the ceramic component. In comparison with pure PDMS, the PDMS–10%PMN–PT allows an increase of more than 60% in the harvested energy per unit volume, while the PDMS–1%PZT composite, entailing a minor improvement, here in the range 23.5–37.4%, exhibits a better performance in terms of generated energy per unit weight. These results provide a guide for the choice and design of materials suitable for the realization of enhanced energy harvesters.

Keywords: Smart materials; Dielectric elastomers; Ceramic fillers; Energy harvesting; Energy generators

1. Introduction

In the recent years, a growing number of efforts within the research community are being devoted to the design of efficient devices for energy harvesting from mechanical work, based on dielectric elastomers (DEs), e.g. Refs. 3, 11, 12, 15. Being reliable, quickly responsive, light, portable, cheap and involving few moving parts, such devices are very attractive towards applications in a variety of fields, i.e. allowing energy extraction from sea waves, water currents, wind, human walking (portable electronics), etc., see e.g. Ref. 4. Dielectric elastomer generators (DEGs), conceived as parallel plate stretchable capacitors with variable capacitance, basically consist of a DE film elastically stretchable coated with compliant electrodes on the upper and lower surfaces. The capacitance depends on the stretch undergone by the DE film (therefore on its area and thickness) and thus it changes as a consequence of stretching and strain relaxation induced by the interaction with the external environment, therefore allowing the extraction of electric energy.

Different configurations and harvesting cycles are being proposed in literature, essentially involving three basic charging–discharging strategies as mentioned in Ref. 3, assuming that along both the stretching and contracting phases induced by the external mechanical force, one candidate among voltage, charge, electric field is kept constant. In the present paper we focus on the variable capacitor described above subjected to a sequence of electromechanical four-step cycles based on the constant charge strategy. In particular, the cycle layout is the following: (*i*) stretching under growing mechanical load and constant electric charge (open circuit condition); (*ii*) electric charge deposition through application of a voltage under constant load; (*iii*) strain relaxation under decreasing mechanical load and constant electric charge (a second open circuit condition), with the voltage increasing correspondingly; (*iv*) electric charge removal and storage under constant load; a net amount of electrical energy is released within this phase over a single cycle.

A major drawback limiting the energy conversion efficiency of DEGs stems from the poor electromechanical coupling characterizing DEs, as indicated by the rather low relative permittivity $\epsilon_r = \epsilon/\epsilon_0$ (being ϵ_0 the permittivity of vacuum, $\epsilon_0 = 8.85$ pF/m) of such materials (generally ϵ_r is few units, between 3 and 7). Nowadays several research projects are

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devoted to the development of enhanced materials, matching the characteristic great deformability of elastomers (low elastic modulus and large elongation ratios) with an improved electromechanical coupling (high permittivity), while also maintaining a high electrical breakdown strength. As mentioned in Ref. 6, single composition materials cannot meet this challenge, while the combination of dissimilar materials is expected to be an effective way to produce high permittivity soft composites, as demonstrated both experimentally[8,17,19,23] and theoretically.[1,9,18,21,24]

The paper focuses on the performance of DEGs made up of composite materials based on an elastomeric matrix with small amounts of ceramic fillers,[a] investigating the possible improvement in terms of specific harvested energy compared to the homogeneous case assuming the same intensity for the external force. Two different materials, recently presented in the literature for different applications (DE actuators, vibration dampers), are considered for the realization of a DEG, both assuming a matrix in silicone dielectric elastomer – PDMS – and a filler in ferroelectric ceramic particles – PMN–PT and PZT. While contributing to an increase of the dielectric permittivity in the composite material, the ceramic inevitably affects the mechanical properties (increasing the stiffness and reducing the ultimate strain) and decreases the dielectric strength of the matrix, thus great attention should be paid in order to find a convenient compromise. Assuming conservative processes and neglecting the lifetime of the device, the performance of generators based on these materials is predicted on a theoretical basis, adopting an optimization procedure developed by the Authors (see Ref. 2) and the maximal harvested energy is evaluated, taking into account the possible failure mechanisms, corresponding to buckling, electric breakdown, excessive deformation and electromechanical instability.

2. DEG's principle of operation and its failure mechanisms

We consider a lossless ideal soft dielectric elastomer film with no volume charge or force, whose points $\boldsymbol{x}$ occupy in the current configuration a parallelepiped-shaped domain $B \in \mathbb{R}^3$ with thickness h along direction x_2 and area A along the orthogonal x_1x_3 plane. The lower and upper surfaces of B, defined as $x_2 = 0,\ h$ respectively, are coated with compliant electrodes, as pictured in Fig. 1, that can be connected to external batteries. The material is incompressible and undergoes large deformations starting from the reference, natural, configuration B^0, so that all the current variables describing the electromechanical problem (stress, charge, electric field, etc.) can be transformed into the corresponding nominal variables with reference to B^0 through pull-back operations (see Refs. 1, 16 and Appendix A).

Neglecting the fringe effects, the superposition of uniform mechanical and electric fields is considered on the body, in particular:

- equal biaxial forces in the midplane represented by the nominal (first Piola–Kirchhoff) total stress components $S_{11} = S_{33} = S$, with $S_{22} = 0$, inducing a state of equibiaxial deformation with in-plane stretch λ, so that the deformation gradient $[F_{ij}]$ assumes the diagonal form

$$[F_{ij}] = \begin{bmatrix} \lambda & 0 & 0 \\ 0 & \lambda^{-2} & 0 \\ 0 & 0 & \lambda \end{bmatrix} \tag{1}$$

  with respect to the orthonormal basis associated with the coordinate system x_1, x_2, x_3, being $J = \det[F_{ij}] = 1$ for incompressibility;
- an electric field $[E_i] = \{0,\ E,\ 0\}^T$ or an electric displacement field $[D_i] = \{0,\ D,\ 0\}^T$ across the thickness, according to the controlled quantity assumed on the electrodes, i.e. the voltage between the two electrodes ϕ or the charge on them Q, respectively, as $E = \phi/h$ and $D = \omega$, where $\omega = Q/A$ represents the charge density. We note that for the corresponding nominal quantities (see Appendix A), $E^0 = \phi/h^0$ and $D^0 = \omega^0$.

The material is assumed to be hyperelastic, described by the extended neo-Hookean strain energy $W^{NH}(F_{kl},\ D_k^0)$

$$W^{NH} = \frac{\mu}{2}(F_{kl}F_{lk} - 3) + \frac{1}{2\epsilon}F_{ij}D_j^0F_{ik}D_k^0, \tag{2}$$

where D_k^0 is the nominal electric displacement and the permittivity ϵ turns out to be independent of the deformation.

Under these hypotheses, the application of the electroelastic constitutive equations (see Appendix A) provide the following relationships in terms of total nominal stress and nominal electric field

$$S = \mu(\lambda - \lambda^{-5}) - \frac{(D^0)^2}{\epsilon\lambda^5}, \quad E^0 = \frac{D^0}{\epsilon\lambda^4}, \tag{3}$$

where the latter corresponds to the well-known equation valid for ideal elastomers, $E = D/\epsilon$, written in terms of current quantities. Note also that for our soft capacitor $(3)_2$ can be alternatively formulated as

$$\phi = \frac{h^0\omega^0}{\epsilon\lambda^4}. \tag{4}$$

The operating principle of the DEG is based on its variable capacitance; this parameter represents a property of the capacitor, defined as $C = Q/\phi$, i.e.

$$C = \frac{\epsilon A}{h} = \epsilon\frac{A^0}{h^0}\lambda^4 = \epsilon\frac{V}{(h^0)^2}\lambda^4, \tag{5}$$

where V denotes the constant volume of the DE film. Similar expressions with different powers of λ hold in the case of different deformation states (e.g. uniaxial or pure shear), therefore the dependence of the capacitance of the DE film on deformation is evident.

More harvesting strategies are available, according to the control parameters adopted along the typical four-phase cycle:[10] we focus on a cycle where either the stress S, representing the effect of the force provided by the external environment onto the

[a] As suggested in Ref. 23, both conductive and insulating fillers can be suitable, but the latter seem to be preferable in order to prevent consistent dielectric losses.