

Electroactive Polymer Composites as a Tactile Sensor for Biomedical Applications

GeunHyung Kim

Samsung Electronics Co., LTD., San#24 Nongseo, Giheung, Yongin, Gyeonggi 449-900, Korea

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Abstract: Modern applications could benefit from multifunctional materials having anisotropic optical, electrical, thermal, or mechanical properties, especially when coupled with locally controlled distribution of the directional response. Such materials are difficult to engineer by conventional methods, but the electric field-aided technology presented herein is able to locally tailor electroactive composites. Applying an electric field to a polymer in its liquid state allows the orientation of chain- or fiber-like inclusions or phases from what was originally an isotropic material. Such composites can be formed from liquid solutions, melts, or mixtures of pre-polymers and cross-linking agents. Upon curing, a "created composite" results; it consists of these "pseudofibers" embedded in a matrix. One can also create oriented composites from embedded spheres, flakes, or fiber-like shapes in a liquid plastic. Orientation of the externally applied electric field defines the orientation of the field-aided self-assembled composites. The strength and duration of exposure of the electric field control the degree of anisotropy created. Results of electromechanical testing of these modified materials, which are relevant to sensing and actuation applications, are presented. The materials' micro/nanostructures were analyzed using microscopy and X-ray diffraction techniques.

Keywords: self-sensing, health monitoring, smart material, polymer composite, microstructure, electrostriction.

Introduction

Polymers are lightweight, tough, and can be easily manufactured into complex shapes, and possess remarkable chemical resistance and biomedical compatibility. Many applications will benefit from multifunctional materials which combine desirable mechanical, thermal and electroactive responses. For example, one can envision a material whose elastic, thermal or electrical properties are locally optimized to enhance its performance, e.g., reduce concentration of mechanical or thermal stresses, ensure desirable heat conduction or dielectric strength. Strong electrostriction response of polymers and polymer composites advances them as candidate for sensors and actuators. Strains as large as 3~100% have been reported for polymers and polymer composites.¹⁻³ Engineering materials with self-sensing capabilities are needed for health monitoring. Combination of adjustable elastic properties with sensing and actuations provide new design approaches for active damping of vibrations and tactile sensors in biomedical applications. Polymeric composites can be locally tailored to optimize material microstructure, composition and morphology, which could be called "a material designed for functions", for various applications

including implantable materials and/or biomaterials using technology which one call electric field-aided micro-tailoring.⁴

One can apply an electric field to make orthotropic composite by forming chain-like structures from spherical inclusions or by orienting embedded flakes or fiber-like particles in a liquid polymer. A solid orthotropic composite results from curing the created structure. The direction of the externally applied electric field dictates orientation of the created composite. Electric field strength and exposure duration control the degree of produced anisotropy. The electric field can be applied to (a) produce a composite with uniformly oriented structure, (b) modify only the surface of manufactured components and (c) locally modify material in selected areas.⁴ Either an AC or DC field can be used to assemble/rearrange organic or inorganic particles whose electric properties differ from those of the original continuous dielectric medium. Moreover, the frequency of the applied field can be chosen such that virtually ANY inclusion can be arranged (or rearranged) in ANY continuous dielectric.⁴ Arranging piezoelectric inclusions in chains may increase actuation properties of the composite.^{5,6} An electric field can be used to form structures with very complex morphology.⁷ Solid electrorheological (ER)^{8,9} and magnetorheological (MR)¹⁰ materials are created by using field-aided technology. Modifying material structure also dramatically affects its electrostriction response. For example, strains up

*e-mail: xrdghk@yahoo.com

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to 117% in silicone elastomers, and up to 215% in acrylic elastomers were observed using biaxially and uniaxially pre-stretched films compared with 30~40% for non-stretched films.² Similarly, appropriate arrangement of inclusions in anisotropic structure will dramatically affect electrostriction response of the material.^{11,12}

The objective of this paper is to demonstrate the feasibility of using an electric field to modify structure of several polymeric composite systems which are formulated by epoxy, silicone and polyamide composites with ceramic, glass fiber, iron and nanoclay, so as to improve their mechanical, thermal, electric and sensing properties.

An orthotropic structure is created by applying electric field to liquid suspension in a polymer. Rotational behaviors of glass fiber in liquid epoxy applied by electric field are shown and the times required to orient are compared with estimated one. The nano-clays in melted polyamide are exposed by electric field and the degree of orientation of particles is measured by using X-ray diffraction. Thermoelastic stress tests are also conducted to show performance of electric field processing for nano-clay/polyamide composites. The measured stress distribution of the composite exposed by electric field is different to that of not applied composite.

Another objective of this research is to evaluate electrostriction sensing responses of the tailored composite, ceramic/silicone system, having anisotropic particle's microstructures. Electrostriction and electrorheological testing of ceramic-silicone composites is conducted for materials with random structure of inclusions, chain-like orthotropic structures and arch-like structures (representing in-plane transverse isotropy). It is shown that both electrostriction and ER-response are manifestations of field-induced stresses and can be described by the same set of material parameters. These parameters, called *electrostriction parameters*, describe deformation-induced changes in dielectric properties of the material. Only two parameters are required to describe field-induced stresses in isotropic material, whereas up to six electrostriction parameters may be needed for simple orthotropic materials. A simple microscopic analysis^{11,12} relates these parameters to material microstructure and predicts that arrangement of inclusions in anisotropic structures affects electroactive response compared with randomly distributed inclusions. Results show that electroactive responses vary with inclusions structures in a matrix material.

Electric Field Aided Microtailoring Technology

One constantly seeks multifunctional electroactive materials. For example, "smart" materials for active vibration damping need sensing capabilities and ability to provide electrorheological response, preferably both in the same material. Other applications such as material health moni-

toring will also benefit from self-sensing capabilities. Anisotropic materials are needed for such applications. Moreover, it is even much more important to be able to produce such anisotropy locally and at will. In this section the electric field aided technology is discussed to fabricate such materials possessing desired microstructure. In section 3 contains measured electroactive properties of tailored ceramic/silicone rubber composites.

By applying an electric field to a polymer in its liquid state, orientated chains or fiber-like inclusions or phases are created from what was originally isotropic material. Such composites can be formed from liquid solutions, melts, or mixture of pre-polymer and cross-linking agent. Upon curing, a "created composite" results which consists of these "pseudo fibers" embedded in a matrix. One can also create orientated composites from embedded spheres, flakes or fiber-like shapes in a liquid plastic. Application of a non-uniform field will cause spatial re-distribution of inclusions. The direction of the externally applied electric field defines the direction of anisotropy of the resulting solid composite. The electric field can be applied to (a) produce a composite with uniformly oriented structure; (b) modify only the surface of manufactured components; and (c) locally modify material in selected areas. Either an AC or DC field can be used to assemble, rearrange and orient organic and inorganic particles whose electric properties differ from those of the suspending medium. The frequency of the applied field can be chosen such that virtually any inclusion can be manipulated in any liquid dielectric. Whereas an applied electric field is used with dielectric polymers, a magnetic field can be similarly used to assemble/rearrange magnetic particles (e.g., iron) in nonmagnetic media, or nonmagnetic particles in magnetic media. Resulting composites have superior mechanical, thermal or electric properties in preferable directions, and exhibit advantageous electrorheological and magnetorheological types of behavior.^{4-6,9,11}

In many ways the so-created composites resemble more traditional discrete-fiber reinforced materials, Figure 1. However, instead of typically being restricted to having fixed effective fiber content and fiber orientation(s) throughout a component, one can now form fiber-like chains of particles ("pseudo fibers") of desired orientation and effective fiber content, and in desired places or regions of an engineered component. Several mechanisms influence the composite structure: (a) dipole-dipole attraction, (b) field-induced torque and (c) spatial redistribution of inclusions in non-uniform applied field.

Consider a material composed of identical polarizable spheres in a dielectric liquid, Figure 1(a). Spherical inclusions move under dipole-dipole attraction^{14,15} but experience a resistance force given by Stoke's law¹⁶ $F = 3\pi \cdot \eta \cdot d \cdot v$, where η is the fluid viscosity and v is velocity of the inclusion. One can estimate the time required to form a chain of inclusions in an electric field by assuming that to do so each

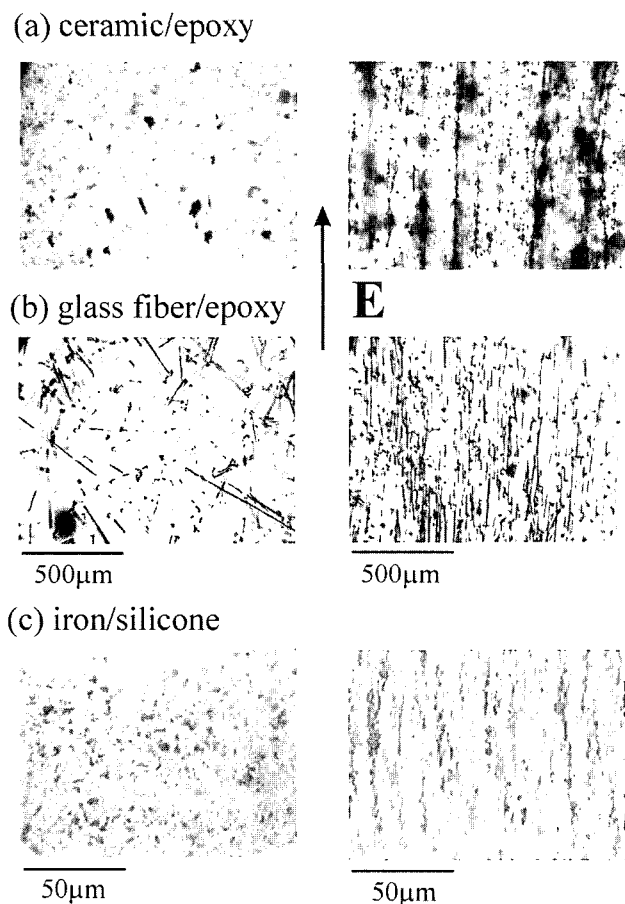


Figure 1. Aligning (a) random ceramic particles in epoxy into “pseudo fiber” structure, (b) randomly oriented glass fibers in epoxy into orthotropic structure by electric field, and (c) tailored composites with iron particles (5 vol%) in silicone rubber.

inclusion moves a distance equal to its diameter, $d = v \cdot t$. The duration of time, in seconds, is proportional to the fluid viscosity, η , in units of Pa.s and the electric field, E , in units of V/m, i.e., $t^{chain} \approx 10 \cdot \eta / \epsilon_0 E^2$.⁴ A similar analysis for fiber-like inclusions gives $t^{fiber} \approx 10^2 \eta / \epsilon_0 E^2$.⁴ A typical value for the electric field strength does not require expensive equipment. Based on the above equations, estimated field-aided processing times required to arrange inclusions in typical engineering plastics are given in reference (4). The rotational behaviors of glass fiber in liquid epoxy are indicated in Figure 2. Data points in this figure were acquire by analyzing individual frames of a high-resolution video and tracking orientations of selected fibers with time. Figure 2(a) shows glass fiber rotations for two different viscosities (= 25 and 75 poise) of continuous phase, liquid epoxy, and various length/diameter (L/D) ratios of fibers. The result shows the fiber’s L/D ratios don’t affect the rotational behavior of the inclusion. The rotational time of the similar size fiber in the liquid epoxy having high viscosity (75 poise) is much longer than that of the particles in the continuous medium having low

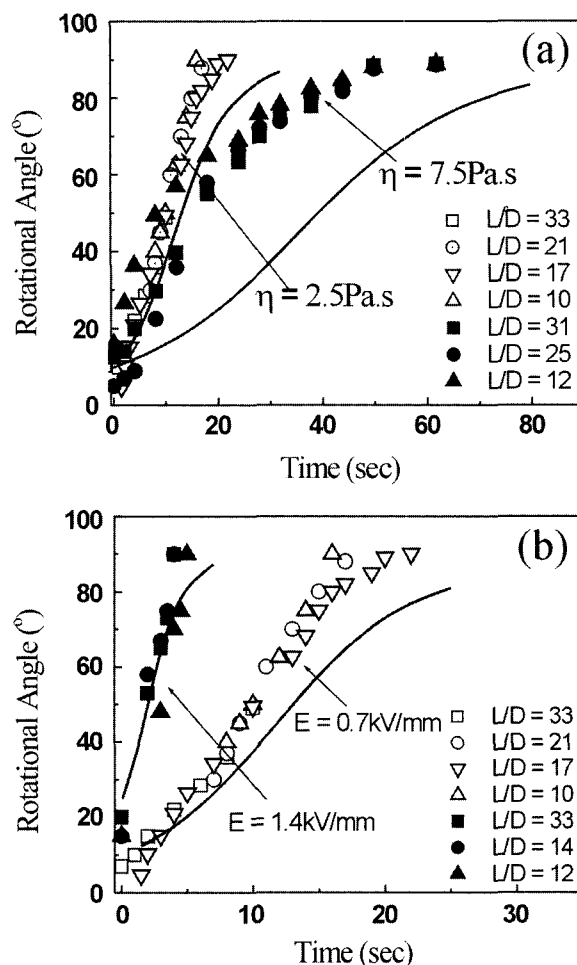


Figure 2. Measured rotational angle vs. time for glass fibers dispersed in epoxy. Solid lines represent theoretical predictions. Symbols are measurements for fibers of different aspect ratio L/D , electric field strength and shear viscosity. Applied electric fields (0.7 kV/mm and 1.4 kV/mm) are well below the dielectric strength, which is in range of 70 kV/mm to 140 kV/mm. (a) $\omega = 5$ Hz, $E = 0.7$ kV/mm, $\phi = 5$ vol% and (b) $\omega = 5$ Hz, $\eta_{epoxy} = 25$ poise, $\phi = 5$ vol%.

viscosity (25 poise). Figure 2(b) describe the rotational behaviors of glass fibers for two different electric fields (= 0.7 and 1.4 kV/mm). At high field strength, the time rotating fibers in a liquid media to the field direction is shorter than those under the low field strength, which is consistent with the estimation of equation, $t \approx 10^2 \eta / \epsilon_0 E^2$. Theoretical predictions in Figure 2 were obtained using balance of field-induced, Stokes and random-oscillatory torques. As seen in Figure 2, there are some differences between measured and predicted curve. However, the oversimplified model was intended to understand major parameters, viscosity and electric field, in this technology. The detail other factors were discussed in reference (4).

Samples of nano-clay/nylon composite (from *RTP Com-*

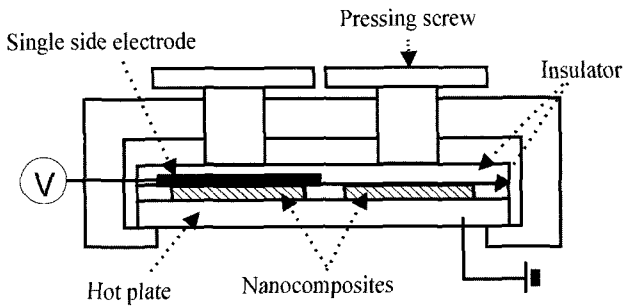


Figure 3. Setup to fabricate the tailored nano-clay/polyamide composite.

pany) having rectangular shape were made by injection molding machine. The obtained samples were cut into two specimens, melted at 250°C and exposed to an electric field for 20 and 40 min in a specially designed mold, see Figure 3. Detail setup is described in the Figure 3. The tailored clay-polyamide nanocomposites were scanned by XRD (X-ray diffractometer) to compare the difference structure between composite exposed to the field and not exposed one. The XRD (Scintag PadV X-ray Diffractometer) used in this testing had a Bragg-Brentano parafocusing geometry. Cu K α ($\lambda = 1.541 \text{ \AA}$) radiation that was generated at a voltage of 35 kV and current of 40 mA was used as the X-ray source. Diffraction angle (2θ) was scanned from 2 to 30° at a rate of 2°/min.

Figure 4 illustrates aligning nano-clay particles by electric field processing. The increased diffraction maximum at $2\theta = 10.5^\circ$ of Figure 4 shows that the electric field has oriented the nano-clay particles. A decrease in the diffraction maximum corresponding to the α -crystals, and increase in diffraction

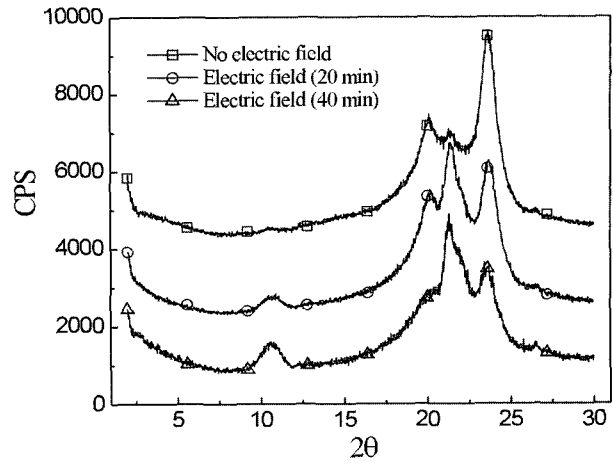


Figure 4. X-ray analysis of silicate clay nanocomposite demonstrates ability of field-aided technology to orient, by applying electric field, both the nanoclay particles (i.e., diffraction maximum at $2\theta = 10.5^\circ$) and the polyamide crystals.

maximum corresponding to the γ -crystals, illustrate that the applied field also affects the crystal structure of the polyamide.

The nature of structural stiffness (compliance) in the neighborhood of geometric discontinuities can affect the stresses, hence strength. It has been well recognized that reducing the amount of material can sometimes actually reduce stress concentrations in isotropic materials.¹⁷ For example, introducing auxiliary holes in wood tensile members reduced stress concentration and increased strength. While drilling holes might be one approach to reduce stiffness, holes are often unacceptable. On the other hand, suitably changing

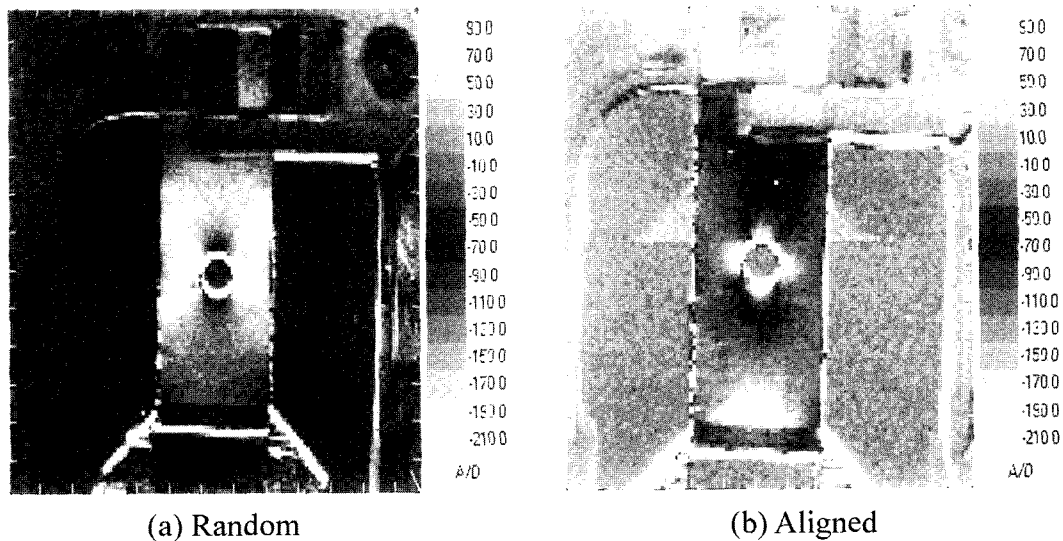


Figure 5. Thermoelastic stress analysis of perforated, loaded nano-clay/polyamide composite (the testing conditions were frequency = 10 Hz and strain = 1% by DMA-2980): (a) randomly distributed inclusions and (b) distribution modified by electric field during 20 min.

material stiffness in appropriate regions by electric field processing could become the solution of choice.

Figure 5 is thermoelastic stress distribution¹⁸ of loaded, perforated nano-clay/polyamide composites that were fabricated from Figure 3. Figure 5(a) is a composite having randomly distributed inclusions, whereas the nano-clays of Figure 5(b) were oriented by an electric field during 20 min. These thermograms indicate ability to modify nano-clay/polyamide composites using field-aided technology and thereby change the stresses around the hole.

Background of Electroactivity

Electrostriction and ER Manifestation of Field-induced Stresses. All materials undergo dimensional change when subjected to an electric field. In general, the total electric field-induced strain, u_{ik} , has both linear and quadratic contributions, $u_{ik} = d_{ijk}E_k + q_{ijkl}E_kE_l$. The first term, which is linear with the magnitude of the electric field, is a manifestation of the *piezoelectric* phenomenon. The second term, which is quadratic with the field magnitude, is the *electrostriction* effect. Piezoelectricity occurs only in some anisotropic materials, but the electrostriction effect exists in all materials. It should be recognized, however, that since deformation depends on the conditions at the material's boundaries, electrostriction coefficients, q_{ijkl} , are not a material property in the strictest definition. Parameters q_{ijkl} measured for one particular set of conditions may have different values under a different set of conditions.¹³ Thus, following Stratton¹⁴ and Landau and Lifshitz,¹⁵ another approach to electrostriction has been proposed.^{12,13} It has been demonstrated that electric field induced stresses are determined by the same material parameters which determine the variation of the material polarization with deformation. A thermodynamic consideration relates field-induced stress, σ_{ik} , to the dielectric displacement, D , and the electric field, E , by

$$\sigma_{ik} = \sigma_{ik}^{mech} - \frac{1}{2} \frac{\partial D_l}{\partial u_{ik}|_{T,E}} \cdot E_l - \frac{\mathbf{E} \cdot \mathbf{D}}{2} \delta_{ik} + \frac{E_i D_k + E_k D_i}{2} \quad (1)$$

where total stress, σ_{ik} , consists of elastic stress, σ_{ik}^{mech} , electrostriction stress, $-1/2 \partial D_l / \partial u_{ik}|_{T,E} \cdot E_l$, and the Maxwell's stress, $-E \cdot D \delta_{ik} / 2 + (E_i D_k + E_k D_i) / 2$. Material parameters describing the effect of strain on polarization, $\partial D_l / \partial u_{ik}$, are called *electrostriction parameters*. Note that by modeling deformation effects on the dielectric displacement, D , for materials of various microstructures and anisotropies, one can evaluate field-induced stresses. This procedure for isotropic and transverse isotropic composites will be implemented in next sections, respectively. Material anisotropy dictates the number of the parameters required to describe field-induced stresses. For example Section 3.2 shows that stresses in isotropic materials can be predicted using dielectric constant, ϵ , and two electrostriction parameters. On the other hand,

electrostatic stresses in transversely isotropic materials depend on two dielectric constants and six electrostriction parameters, see Section 3.3.

The two phenomena, electrostriction and electrorheology, can be described using the same approach. Electrostriction deformation is due to field-induced stresses and occurs in all materials. Electrorheology (ER), which is defined as the field-induced change in rheological properties, is typically only observed in anisotropic materials. On the other hand, the electrostriction effect, defined as changing material polarization with shear or normal strains, offers new sensing approaches.^{9,19} Conventional capacitance sensing techniques possess intrinsic limitations, such as low dynamic range, limited microscaling, complexities in micromachining. Electrostriction technology, while resembling traditional capacitance sensing technique, overcomes most limitations of the former.²⁰ Variation in dielectric properties with deformation (electrostriction) is a fundamental property of any dielectric material. Therefore, a large selection of materials is available for electrostriction sensing media. One feasible sensor design would use the material being monitored as the sensing medium to provide non-invasive material health monitoring and to form "smart" material having self-sensing capabilities. Interdigitated electrodes can be directly embedded into the component of interest, located between plies of a laminate or attached to the surface. Self-sensing capabilities of a component can be enhanced by locally modifying its structure. Electrostrictive materials possess a tunable aspect which could potentially make their response much stronger than that of piezoelectrics. Typical engineering materials are much more compliant than ceramics which are traditionally used in *piezoelectric sensors*. For the same loads, engineering materials experience much larger deformations and produce a very strong output voltage signal. For example, a 100 μm thick plate of PZT4D (Lead Zirconate Titanate) will generate 0.25 V potential under a pressure of 10^5 Pa (corresponding to 1 atm). A polyurethane elastomer of Young's modulus $E_y = 10^6$ Pa and relative dielectric constant $\epsilon = 6.5$ provides a similar sensitivity for a voltage bias of only 3 V. Such a small bias voltage can be readily supplied by the same electronics that provides signal conditioning.

Electrostriction of Isotropic Materials. Isotropic dielectric material becomes anisotropic after deformation and their dielectric properties should then be described by a second rank dielectric tensor, ϵ_{ik} ,^{14,15}

$$\epsilon_{ik} = \epsilon \delta_{ik} + \alpha_1 u_{ik} + \alpha_2 u_{ll} \delta_{ik}, \quad (2)$$

where ϵ is the dielectric constant of the undeformed material, δ_{ik} is the unit tensor, and α_1 and α_2 are electrostriction constants. Note $u_{ll} = u_{11} + u_{22} + u_{33} \approx \Delta V / V$ represents relative change in a volume with deformation. Parameter α_1 describes the effect of strain on the dielectric properties of the material, and α_2 relates the material's dielectric response to relative

change in volume. Eq. (2) relates deformation to material's dielectric properties for isotropic materials. Combining Eqs. (1) and (2) gives the field-induced stress in terms of electrostriction parameters, α_1 , α_2 , and dielectric constant, ε , as follows,

$$\delta_{ik} = \frac{1}{2} \varepsilon_0 [(2\varepsilon - \alpha_1) E_i E_k - (\varepsilon + \alpha_2) E^2 \delta_{ik}], \quad (3)$$

where $\varepsilon_0 = 8.85 \times 10^{-12} \text{ F/m}$ is the permittivity of free space. Integrating $\sigma^{elect.}$ over the surface of the body gives total electrostatic force. Eqs. (2) and (3) show interconnections between two definitions of electrostriction. One can see the same material parameters, α_1 and α_2 , define relationship between dielectric properties of deformed material in Eq. (2) and define field-induced stresses in Eq. (3). A theoretical model for parameters α_1 and α_2 predicts them to be proportional to ε^2 . Parameters α_1 and α_2 can also be obtained from dielectric measurements under constrained and unconstrained sample.¹³ For example, the variation of dielectric properties with thickness change, $\Delta h/h$, of constrained dielectric material is $\Delta\varepsilon = [\alpha_1 + \alpha_2] \Delta h/h$. Similarly, variation of dielectric properties with thickness change, $\Delta h/h$, of unconstrained dielectric material is $\Delta\varepsilon = [\alpha_1 + (1 - 2\nu)\alpha_2] \Delta h/h$, where ν is the Poisson's ratio of the material. Electrostriction measurements for both isotropic and anisotropic materials were shown in Experimental. Note that Eq. (3) predicts no shear stresses in planes perpendicular to the applied field. This result is in agreement with experimental data presented in ER test. Sensor applications of electrostriction measurements are discussed in Figure 8, and elsewhere in more details.²⁰

Electrorheology and Electrostriction of Transversely Isotropic Materials. An important type of anisotropy, called *transverse isotropy*, has a single distinct direction which can be represented by the unit vector d .¹² Electrorheological suspensions and elastomers, where particle columns or chains are oriented in a common direction, are typical examples of transversely isotropic materials. The dielectric properties of transversely isotropic materials differ in the d direction from those the plane perpendicular to d . These materials have a second order dielectric tensor of the form $\varepsilon_{ik} = \varepsilon^{\parallel} \delta_{ik} + (\varepsilon^{\perp} - \varepsilon^{\parallel}) d_i d_k$, where ε^{\parallel} and ε^{\perp} are the relative dielectric constants associate with the fields applied parallel and perpendicular to d , respectively. Overall six *electrostriction coefficients*, α_1 through α_6 , are required to predict field-induced stress and the effect of deformations in transversely isotropic materials. These material properties are scalar functions of thermodynamic variables such as the temperature, the electric field magnitude and the density. In some common situations, e.g. normal and shear deformations, fewer than six parameters are needed. The effect of deformations in the *transverse direction* and the *normal stress* in that direction are^{9,12}

$$\Delta\varepsilon = \varepsilon - \varepsilon^{\parallel} = K_n u_{33} \quad \text{and} \quad \sigma_{33}^{elect.} = \varepsilon_0 \frac{E^2}{2} [\varepsilon^{\parallel} - K_n], \quad (4)$$

where deformation effect on polarization and normal stress are both determined by the same parameter K_n . If the material is subjected to *simple shear* with the displacement field $U = (0, \gamma x_3, 0)$, strain components are $u_{13} = u_{31} = \gamma/2$. The effect on dielectric properties, $\Delta\varepsilon$ (shear electrostriction) and shearing stress in electric field $\sigma_{13}^{elect.}$ (electrorheology) are now both described by the same parameters, K_n and K_{τ} .^{9,12}

$$\Delta\varepsilon = (K_n + K_{\tau}) \frac{\gamma^2}{2} \quad \text{and} \quad \sigma_{33}^{elect.} = \varepsilon_0 \frac{E^2}{2} K_{\tau} \gamma, \quad (5)$$

A theoretical model of isolated chains¹¹ predicts parameters K_n and K_{τ}

$$K_n = -(\varepsilon^{\parallel} - \varepsilon_c) \left[1 + \frac{3\xi}{1-\xi} \right], \quad K_{\tau} = (\varepsilon^{\parallel} - \varepsilon^{\perp}) + (\varepsilon^{\parallel} - \varepsilon_c) \frac{\xi}{1-\xi}$$

$$\text{and} \quad \xi = \frac{1}{2} \frac{\varepsilon_p - \varepsilon_c}{\varepsilon_p + 2\varepsilon_c} \zeta(3) \left(\frac{a}{l} \right)^3, \quad \zeta(3) = 1.202\dots, \quad (6)$$

where ε_p and ε_c are dielectric constants of inclusions and continuous phase, a is the particle diameter and l is the distance between centers of particles in the chain.

Experimental

Materials. An uncured silicon liquid (from Silicons, Inc) was mixed with 10 vol% of ceramic powder (Zeeosphere™ Ceramic Microspheres from 3 M Corp.) and de-gased in a vacuum chamber. With added cross-linking agent, the mixture was evacuated for 5 min and transferred between parallel plates (1 mm gap) of a capacitor. A composite with aligned particles was created by applying frequency = 100 Hz during curing. A composite with randomly distributed ceramic particles, Figure 6(a), was obtained in the absence of an electric field. An arch-like structure was produced by applying a high voltage potential of 1.0 kV across an interdigitated system of electrodes. Similarly, a chain-like structure of particles, Figure 6(b), is formed by applying 4.0 kV across an additional top electrode and one of the sensor's electrodes.

Experimental Setup and Testing Procedures. The interdigitated electrode pattern on the surface in Figure 7 was created by wet etching photosensitive copper-coated plates from Kaypro Electronics. The electrodes of the single-plane capacitance sensor are sets of conductive leads of equal width and spacing. Two sensor elements having electrode patterns configured in two orthogonal directions are needed to detect two components of shear deformation. One sensing element is mounted with its electrodes parallel to the applied displacement, while electrodes of another sensing element are oriented perpendicular to the applied displacement. This

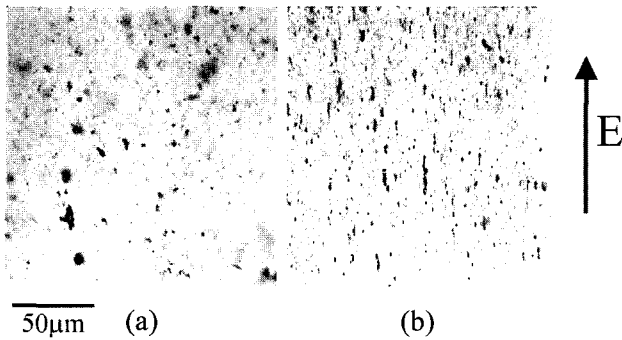


Figure 6. Micrograph of ceramic powder in silicone rubber shows (a) random particles and (b) particles arranged in chain-like structures by electric field.

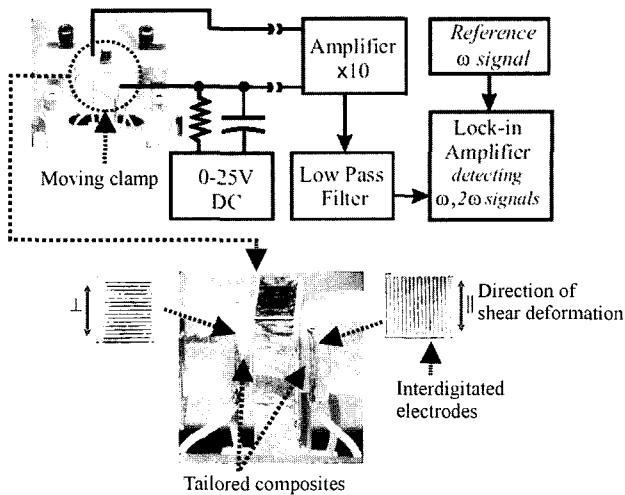


Figure 7. DMA experimental setup showing orientation of electrodes and dielectric media.

solid-state sensor detects deformations through changes in dielectric properties of the attached material. The capacitor utilizes variation in dielectric displacement due to electrostriction, i.e., deformation of the dielectric layer affects the material's dielectric constant and therefore the sensor's capacitance.

The electrostriction measurements with random and aligned structures were performed using DMA-2980. The DMA is able to provide controllable shear deformations of different amplitudes and frequencies which are compared to the sensor response. The rheometer with shear sandwich configuration was modified for electrostriction and electrorheological characterization of samples with aligned and random structures. A static normal force was applied up to 10% oscillatory shear deformation to the rubber composite. Two sensor elements having electrode patterns configured in two orthogonal directions are needed to detect two components of shear deformation. One sensor elements was always kept in the instrument and used as a reference material to adjust the

applied normal force. The electrodes and the actual testing configuration are shown in Figure 7.

A voltage produced by the sample has enormously high source impedance-typically greater than 400 MΩ-which cannot be measured by conventional instrumentation having typical input impedance 1 MΩ, special custom-made conditioning electronics is required. High impedance DC power supply is providing a constant dielectric displacement *D* across the sample. AC signal due to the sample deformation is buffered and amplified by a high impedance amplifier and measured by Stanford Research Systems Lock-in-Amplifier.

To study an ER effect, a high voltage potential in range 0-600 V/mm was generated by a Trek HV amplifier. Electrorheological properties of the sample were acquired at electric field strengths.

Results and Discussion

The output voltage from the two sensing elements strongly depends on material structure, Figure 8. The results can be explained by next expressions. From the Eq. (5), the author can obtain next expressions. Capacitance *C* of the sample and voltage *V* due to charge *Q* are related as $Q = VC$. Variation in the capacitance ΔC with deformation of the charged sample causes variation of the voltage ΔV across the sample

$$\frac{\Delta V}{V} = -\frac{\Delta C}{C}$$

$$\therefore \frac{\Delta V}{V} = -\frac{\Delta \epsilon}{\epsilon} = \frac{1}{\epsilon} (K_{\tau} + K_n) \frac{\gamma^2}{2}$$

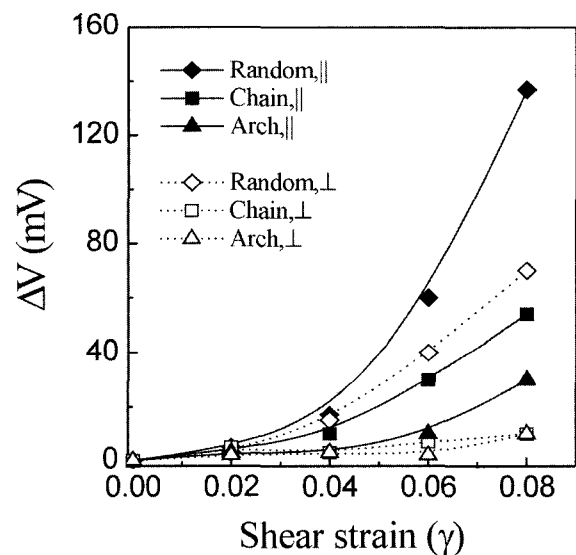


Figure 8. Shear electrostriction sensor with silicone-ceramics composites: the output signals strongly depends on the shearing direction. Solid symbols represent displacement parallel to the electrodes and open symbols represent displacements perpendicular to the electrodes.

According to above equations, variation in capacitance is proportional to the square of the shear strain, γ^2 , which means that oscillatory shear deformation occurring at frequency ω produces a voltage signal ΔV with frequency 2ω . Furthermore, the voltage response must be quadratic with shear magnitude. In Figure 8, the data shows that the second harmonic of the voltage response is quadratic with respect to shear deformation. The response varies with orientation of the sensing element and microstructure of the composite. The voltage response is strongest for the random system and weakest for the arch-like structure. The effect also depends on the shearing direction. The effect is stronger when the shearing is parallel to the electrode direction than when it is perpendicular to the electrode.

In this test, a voltage signal at first harmonic was also observed. This signal appears because the dielectric polymer material used as a sensing media has viscoelastic properties. The total deformation, γ is composed of two contributions, elastic deformation, γ_e , and plastic deformation, γ_p , where $\gamma = \gamma_e + \gamma_p$. Only elastic deformation, γ_e , contributes to the electrostriction effect. Thus for oscillatory shear deformations $\gamma = \gamma_p + \gamma_e \sin \omega t$ and the voltage response, which is proportional to γ^2 will have two harmonic frequencies of oscillation $2\gamma_p \gamma_e \sin \omega t$ and $\gamma_e^2 \sin 2\omega t$.

The silicone-ceramics composites tested for shear electrostriction were also tested for ER response using DMA-2980 with shear sandwich clamps. The electric field is applied across the sample using parallel plate electrodes. One can see in Figure 9 that ER response is strongly dependent on material anisotropy. The effect is strongest for chain-like structure and is almost absent for a random structure. These experimental results are in agreement with analytical predictions in BACKGROUND. While Eq. (3) predicts no ER-

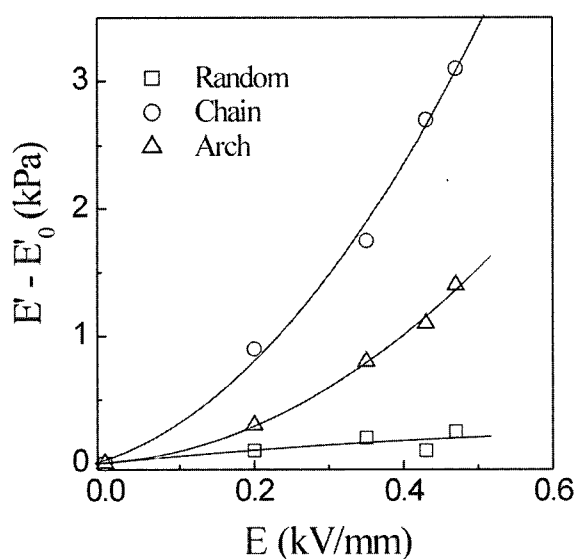


Figure 9. Electrorheological testing of ceramic-silicone composites.

effect for an isotropic composite which has a random structure of inclusions, Eqs. (5) and (6) predict strong effect for transversely isotropic material which is typically represented by composites having chain-like structure of inclusions. It should be noted that the silicone-ceramic composite which was tested has no well-developed structure of the chains. Rather, it has a system of prolonged isolated clusters of particles which are visible in microphotograph of Figure 6(b). This proves that ER effect exists in any transversely isotropic material and is due to field-induced stresses in anisotropic materials but not due to mechanical interactions between particles in chains.¹² Shear electrostriction measurements in Figure 8 cannot be directly compared with ER measurements in Figure 9 because these two measurements have different orientations of electric field with the respect to composite structures.

Conclusions

This study demonstrates ability to modify composites consisting of ceramic, iron, glass fibers and nano-clay platelets in silicone rubber, epoxy and polyamide using electric field. The results show that polymeric composites of various constituents can be engineered and micro-tailored to fulfill desired objectives. The technology is able to locally manipulate the orientation and structure of micro- or nano-sized inclusions in a polymeric matrix to enhance composite performance and optimize functionally graded materials. For example, the locally controlled orientation of carbon nano-tubes (CNTs) which have been used as field emission cathodes, micro heat sink, biomedical materials, and polymer composites with locally thermal or electrical path could be obtained. Microstructures of resulting solid composites are aligned with the applied field. The field can be employed to (a) produce materials having uniformly oriented structures, (b) modify only the surface of a manufactured component, and/or (c) locally modify material in selected regions. The results demonstrate that polymeric components can be modified up to 5 mm in depth with readily available voltage sources and at acceptable processing times. Controlling transversely isotropy is shown here to be capable of modifying electroactive response and thermoelastic properties of materials. Measured results are compatible with theoretical predictions. The technology is also expected to be able to provide self-sensing and "smart" material capabilities. Current effort emphasizes achieving mechanical, actuator and sensor design objectives, although thermal and electrical responses can be similarly treated. Future developments of the technology could also involve space and time variable electric fields (for example, rotating electric field). Such field configurations will enable one to move inclusions in desirable areas within a volume.

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