

Effect of Filler Materials with Different Cross Sections on the Performance of Dielectric Polymer Actuators

Pankaj Kumar Singh, Kamal Sharma, Pradeep Kumar Singh

Abstract: Electroactive polymers with quick response time, suitable for most actuator applications, are broadly classified into three categories: Piezoelectric polymers, Dielectric elastomers and electrostrictive polymers. Among the above three, dielectric elastomers polymers offer the best performance characteristics. It is well known that the underlying microstructure influences the overall properties of a composite. In this regard, our first objective here is to investigate the effect of volume fraction and distribution of high dielectric constant ($K \sim 10000$) filler particles on the overall dielectric constant of EAP composites. The filler particles are taken to be cylindrical with circular cross-sections and the dielectric behavior of the phases is taken to be linear. Both random as well as periodic microstructures are analyzed. More specifically among the periodic microstructures, a square and hexagonal arrangement of cylindrical fibers is considered. In experiments, nano-composite containing very high dielectric constant inclusions is shown to result in very high overall dielectric constant at volume fractions less than 10%.

Keywords: Dielectric elastomers, Dielectric constant, Periodic nano-composite, Mechanical properties.

I. INTRODUCTION

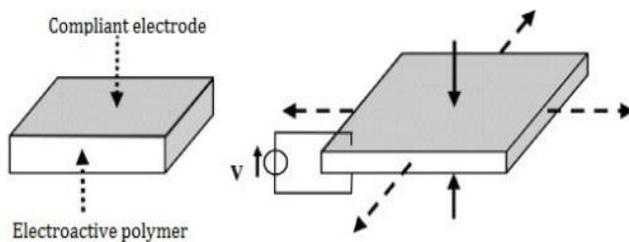


Fig. 1 Mechanical deformation of Electroactive polymer. Right: Voltage is ON; Left: Voltage is OFF. Maximum strain of over 100% have been reported.

Electroactive polymers (EAPs), which undergo shape changes when subjected to electric fields as shown in Fig. 1 [1][2][3][4][5][6], are attractive for designing a wide range of sensors and actuators. In comparison to traditional actuation system based on motors, solenoids, voice coils, piezoelectric ceramics, shape memory alloys, pneumatics and hydraulic, actuators based on EAPs are light, flexible, easy to process, inexpensive, and can be conformed to complicated surface and geometries. [7] Until recently, high operational voltage (on the order of 200 V/ μm) a low

performance characteristics (maximum strain attainable, maximum energy density, and coupling efficiency) limited their use. However, recent breakthroughs in the design of high performance EAPs composite with relatively low operational voltage (on the order of 10 V/ μm) now allow us to develop actuators based on these materials that can compete with conventional electromagnetic and piezoelectric actuators in performance and cost and can be used for developing complex systems such as artificial muscle, miniaturized robots mimicking biological systems (e.g. insects), or biomedical instruments for conducting non-invasive surgeries. [8] Even though, numerous experiments have been conducted to characterize the behaviour of EAP composites, the theoretical understanding of these materials is rather limited. Since, models predicting overall electromechanical response of EAP composites as a function of their microstructure are key to optimal design of EAP actuators, development of such models shall be the main aim of this work. Electroactive polymers with quick response time suitable for most actuator applications are broadly classified into three categories: Piezoelectric polymers, exhibiting electromechanical coupling from the torque exerted by electric field on the molecular dipoles, Dielectric elastomers (synthetic rubbers), exhibiting electromechanical coupling from Coulomb interaction between the charges on the electrodes coated on to the specimen (Maxwell stress effect) [9], and Electrostrictive polymers, deforming when placed in electric field from changes in molecular conformations and phase changes in the crystalline regions. Among the above three EAPs, dielectric elastomers and electrostrictive polymers offer the best performance characteristics and shall therefore be the focus of this work. In particular, the focus shall be on particle reinforced polyurethane (PU) and Poly(vinylidene fluoride-trifluoroethylene-chlorofluoroethylene) (VDF-TrFE-CFE) composites [10] Dielectric elastomers and electrostrictive polymers have been the centre of most experimental and theoretical investigations on EAPs [11][12][13][14] since they offer the best performance characteristics among EAPs. Table 1 compares the performance characteristics of best performing dielectric polymers such as PUs and silicones and best performing electrostrictive polymers such as P(VDF-Tr FE-CFE) with characteristics for natural muscle and the state of the art piezoelectric ceramics such as Lead Zirconate Titanate (PZT) polycrystals and Lead Zirconate Niobate – Lead Titanate (PZN-PT) single crystals. Performance of actuator materials is being measured by a number of parameters including maximum attainable strain, stress, energy density, coupling efficiency, and the operating electric field (further information regarding which is provided in the following).

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EAPs a specified mechanical output per stroke, the coupling efficiency relating the strain energy stored in the actuator to the electrostatic energy stored in the dielectric, determines the efficiency of the actuator (higher the coupling efficiency lower the losses encountered in recovery electronics). Actuator materials would find limited practical use, if their operating voltages are too high (from tens of KV's to MV's). Therefore, electric field at which the maximum performance is observed (shown in the 7th column of the table) is also taken as an actuator parameter. [15][16] Lastly, it must be noted that the performance characteristics are sensitive to operating temperature and that the characteristics shown in Table 1 are evaluated at room temperature (~25°C). The specific objectives of this work are as follows:

- Investigate the effect of size, shape, volume fraction, and distribution of high dielectric constant (K~10000) filler particles on the effective dielectric constant and dielectric strength of dielectric composites. In this work, the inclusions shall be assumed to be ellipsoidal (such that spheres, fibers, and plates form the limiting cases of inclusion shapes), the mechanical response of the polymer matrix and the inclusions (which may be organic or inorganic) shall be taken to be described by hyperelastic potentials, and the dielectric behavior of the phases shall be taken to be linear or paraelectric. Furthermore, both random as well as periodic microstructures shall be analyzed.

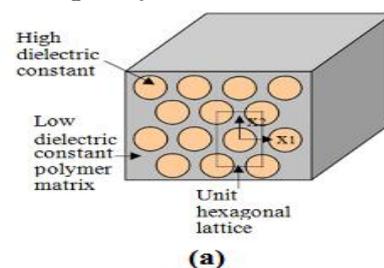
II. PERFORMANCE OF VARIOUS ACTUATORS MATERIALS

Many important observations can be made from Table 1[8][17][18][19][10]. It can be seen that PZTs, the most widely used piezoelectric ceramics, can generate large stresses with reasonably good coupling efficiencies (>50%) at relatively low electric fields (< 5V/μm). However, the deformations they can generate are quite small (the maximum attainable strain is ~ 0.2%). On the other hand, relaxor PT single crystals such as PZN-8%PT, in which hysteresis losses arising from ferroelectric behavior are drastically reduced, exhibit far superior properties. In fact, there is almost an order of magnitude difference in maximum attainable strain and energy density in PZN-PTs and PZTs. However, PZN-PT single crystals are expensive and difficult to fabricate and therefore used mainly only in high performance devices. In contrast to piezoelectric ceramics, the dielectric elastomers such as PU and silicones can exhibit large deformations (with maximum attainable strain up to 40%) with reasonably high energy densities and coupling efficiencies. However, the maximum pressure exerted by dielectric elastomers is relatively small (on the order of 1 MPa) and the operating electric field is quite high (>150V/μm). Interestingly, Silicones exhibit properties that are very similar to those for human muscle. Among Poly(vinylidene fluoride-trifluoro ethylene) P(VDF-TrFE) based electrostrictive polymers, P(VDF-TrFE-CFE) exhibits the best performance characteristics. It can not only exert large stresses (on the order of 50MPa) like piezoelectric ceramics but also undergo considerable deformations like dielectric elastomers (with strains to the tune of 5%). Furthermore, they can achieve high energy densities and can operate with good coupling efficiencies. Nevertheless, like dielectric elastomers they suffer from the drawback that the operating electric field is quite high (>100V/μm). Surprisingly, however, this drawback in dielectric elastomers and electrostrictive polymers can be overcome with ease by

constructing composites of actuator materials with high dielectric constant organic nano-fillers. For example, by constructing a nanocomposite of PU with 3.5 vol % copper phthalocyanine (CuPc), the dielectric constant of the elastomeric system can be increased from 7 to 1000. Consequently, the operating electric field can be brought down from 160V/μm to 11.5V/μm.

III. MODELLING

EAPs such as PUs, Silicones, and P(VDF-TrFE-CFEs) exhibit very good electromechanical coupling characteristics[20]. However, since polymers inherently exhibit low dielectric constants, electric fields required to achieve reasonable actuation pressure or stroke in these materials are quite high. A simple way of reducing the operating electric field in these materials is to increase their dielectric constant by dispersing high dielectric constant nano fillers in them. Although the inclusion of high dielectric constant nano fillers increases the average dielectric constant of a composite, they also produce a highly inhomogeneous electric field with local hot spots that reduce the effective dielectric strength of the composite. Since the electric energy density scales quadratically with electric field, this reduction in breakdown strength could be a serious drawback of the nanocomposite approach. Thus, the main challenge is to come up with composite microstructures that balance seemingly contradictory criteria of enhancing dielectric constant while maintaining high dielectric strength. Two kinds of initial microstructures (initial distributions of phases) are considered in these analyses: periodic and random as shown in Fig. 2(a), Fig. 2(b) and Fig. 2(c). More specifically, two-phase particulate microstructures are considered with periodic and random distribution of ellipsoidal inclusions. While inclusion volume fraction, ellipsoid aspect ratios (spherical, cylindrical and lamellar inclusions would occur as three special cases), and lattice configurations (e.g. simple cubic, body centered cubic, or face centered cubic arrangement of inclusions in three dimensions or square, rectangular, or hexagonal arrangement of cylindrical inclusions in two dimensions) form the microstructural variables in periodic microstructures, n-point probability statistics involving ensemble averages of appropriate functions form the microstructural parameters in random composites. It is important to note that it is not only difficult to extract higher order statistical information from random composites but also equally difficult to use this information in constitutive models. In this work we make use of one- and two-point probability statistics of random microstructures that provide information on the volume fraction and average shape and distribution of inclusions.



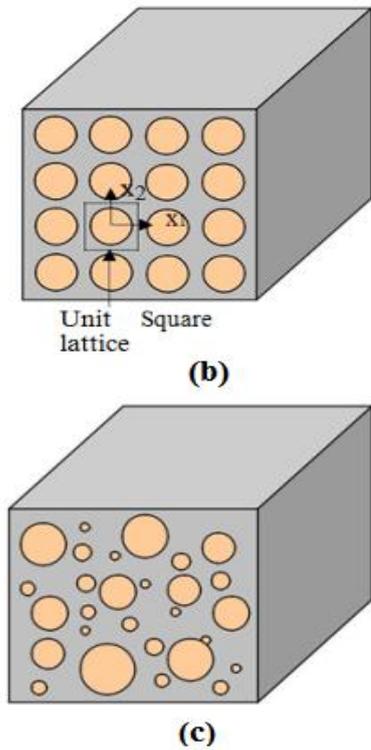


Fig. 2 (a) Polymer composites with hexagonal periodic arrangement of high dielectric constant fibers. Ratio of dielectric constants of fiber to the matrix is taken to be 100. Dashed line shows the unit cell structure. 2 (b) Composite with square arrangement of high dielectric constant fibers. 2 (c) Random composite with cylindrical fibers.

IV. EFFECTIVE BEHAVIOUR OF DIELECTRIC COMPOSITES

The organic fillers commonly used in the dielectric composites exhibit similar mechanical properties as that of the matrix polymers. Therefore, the main aim here shall be to study the effective dielectric properties of dielectric composites as a function of the initial microstructure. Motivated by experimental observations for commonly used dielectrics the dielectric behavior of the polymer matrix as well as the nano fillers are taken to be linear and isotropic such that the local electric flux density $D = \epsilon_0 k E$, where k is the local dielectric constant (note that $k = k_f$ for the nano fillers and k_m for the matrix), ϵ_0 is the electrical permittivity of vacuum, and E is the local electric field.

Thus, while dielectric strengths and linear dielectric constants of the filler and matrix constitute the relevant material properties which are provided as inputs, effective dielectric constant and dielectric strength tensors constitute the relevant macroscopic properties of the composite that we aim to compute. Note that the effective dielectric constant and dielectric strength of a composite can be anisotropic even though the phases are isotropic because of anisotropic distribution of phases (consider e.g. laminated or fiber reinforced composites). Special emphases will be laid on analyzing practically relevant isotropic and transversely isotropic dielectric composites with spherical and cylindrical inclusions. To see how effective dielectric properties can be computed for periodic composites consider the microstructure with hexagonal arrangement of unidirectional fibers. For the choice of reference axes shown in the Fig. Fig. 3(a) and Fig. 3(b), from symmetry considerations, it is easy to show that the effective dielectric constant tensor

K_{eff} (effective dielectric constant) relating with relating the average electric field \bar{E} to the average electric flux density $\bar{D} = \epsilon_0 K_{eff} \bar{E}$ in the composite can be written as

$$[K_{eff}] = \begin{bmatrix} k_{11} & 0 & 0 \\ 0 & k_{11} & 0 \\ 0 & 0 & k_{33} \end{bmatrix}$$

Where k_{11} is the effective in plane dielectric constant and k_{33} is the effective dielectric constant along the fibers. Noting that $\bar{D} = \epsilon_0 k_{11} \bar{E}$ would be the average electric flux density along the x-direction resulting from a unit macroscopic electric field $\bar{E} = 1$ along the positive x-direction, it is evident that the effective in plane dielectric constant can be computed by solving for the average flux density resulting from a unit electric field applied along the x-direction. In addition, since typically the representative volume element of a composite is taken to be composed of a large number of inclusions and since constant potential boundary conditions consistent with uniform electric field of $\bar{E}_{11} = 1$ would be applied on boundary of the composite, the electric field distribution around each of the fibers will be identical. Therefore, the effective in plane dielectric constant can be obtained by solving for the electric flux density distribution $D = \epsilon_0 k E$ (with $E = -\nabla f$ where f is the electric potential) around a single fiber. More specifically, the effective in plane dielectric constant k_{11} can be computed as

$$k_{11} = (\bar{D} / \epsilon_0) = -\frac{1}{A} \left[\int_{\Omega_m} k_m \frac{\partial \phi}{\partial x_1} dA + \int_{\Omega_f} k_f \frac{\partial \phi}{\partial x_1} dA \right]$$

Where A is the area of the unit lattice $W = W_f + W_m$. The electric potential field appearing in the above equation should be such that it satisfies the governing equation for electrostatics:

$$-\Delta \times K \Delta f = 0 \text{ in } W,$$

and

$$\phi(-a, y) = a, \phi(a, y) = -a, \frac{\partial \phi}{\partial y}(x, \pm a) = 0$$

on the boundary. Lastly, noting that for electric fields aligned with the fiber direction, the electric flux density field would be constant per phase, the effective dielectric constant along the fibers becomes

$$k_{33} = c_0 k_f + (1 - c_0) c_0$$

Where c_0 is the fiber (filler) volume fraction. The dielectric breakdown in the composite occurs from the formation of localized zones – running from one electrode to the other - in which the electric (stress) field equals the dielectric strength of the local phase. Thus, the effective dielectric strength is going to be a function of not only the distribution of phases and dielectric properties of the phases but also the orientation of applied electric field with respect to the microstructure.

The dielectric strength \bar{E}_{11}^b for the composite for electric fields applied along the x-direction can be computed by solving for electric potential f such that $\nabla \times D = 0, D = D(-\nabla f)$ in W , and

$$\phi(-a, y) = \bar{E}_{11}^b, \phi(a, y) = -\bar{E}_{11}^b, \frac{\partial \phi}{\partial y}(x \pm a) = 0,$$

on the boundary, and the electric field in the localized zone connecting the two electrodes is equal to the dielectric strength of the phases (E_m^b for the matrix and E_f^b for the fillers), i.e. in the localized zone $\|E\| = E_p^b$ (with $p = m, f$ for the matrix and filler phases, respectively). The relation between the electric flux density D and the electric field $E = -\Delta\phi$ for each of the phases is taken to be of the form. Note that the phase behaviors adopted here are similar to the elastic-perfectly plastic relations in metal plasticity (where the yield stress plays the same role as the dielectric strength over here). Analyses for periodic composites shall be computed using the commercial software package ABAQUS in conjunction with user material models for modeling the elastic-perfectly plastic type response of dielectric materials while random composites can be analyzed using Mori-Tanaka type approximation schemes.

V. RESULT & DISCUSSION

In this work, the attention is limited to polymer composites with inclusions in the form of fibers. Detailed analyses have been conducted for random composites with cylindrical fiber inclusions as well as periodic composites with square and hexagonal arrangement of high dielectric constant fibers as shown in Fig. 2(a), Fig. 2(b) and Fig. 2(c). The fibers are taken to have a dielectric constant 100 times that of the matrix and the effective inplane and out of plane dielectric constants k_1, k_3 , respectively, are computed as a function of volume fraction of the fibers f_0 . The aim here is to analyze the effect of microstructure on effective dielectric constant and to know as to what happens when we reach the percolation limit.

Fig. 2(d) and Fig. 2(e) shows the overall in-plane dielectric constant k_1 as a function of volume fraction of the fibers for hexagonal and square lattices. Also shown in the fig. are effective in-plane dielectric constants for random composites as a function of volume fraction of the fibers. It can be noted that, for periodic composites, the effective dielectric constant increases significantly at the percolation volume fraction which for square arrangement of fibers is equal to $\pi/4$ and for hexagonal arrangement of fibers is 0.907. Furthermore, the Hashin Shtrikhman lower bound (HS LB) for in plane dielectric constant of transversely isotropic microstructures matches very well with the FEM predictions for hexagonal lattices. At lower volume fractions, i.e. at volume fractions considerably smaller than the percolation limits, it is evident that both random and periodic microstructures give identical estimates for effective in-plane dielectric constant.

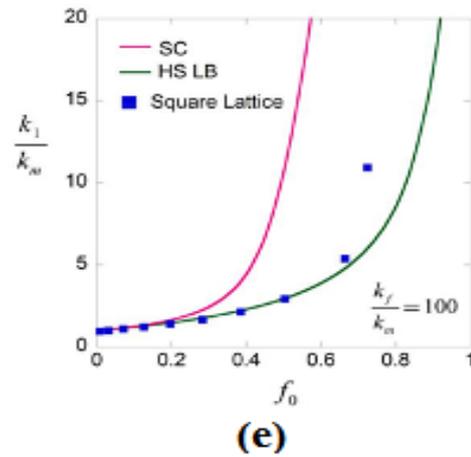
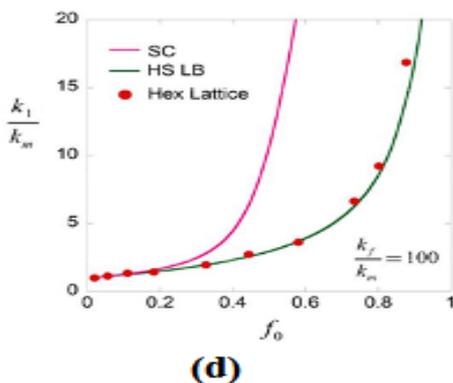


Fig. 2 (d) Effective in plane dielectric constant for periodic composites with hexagonal lattice structures as a function of fiber volume fraction. Also shown are Hashin Shtrikhman lower bound (HS LB) and self-consistent (SC) estimates for random composites. (e) Effective in plane dielectric constant for periodic composites with square lattice structures as a function of fiber volume fraction. Also shown are Hashin Shtrikhman lower bound (HS LB) and self-consistent (SC) estimates for random composites.

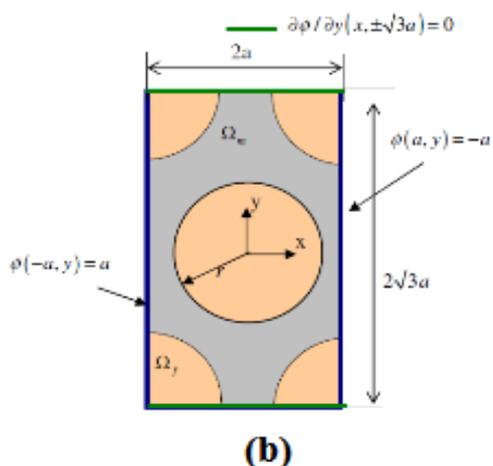
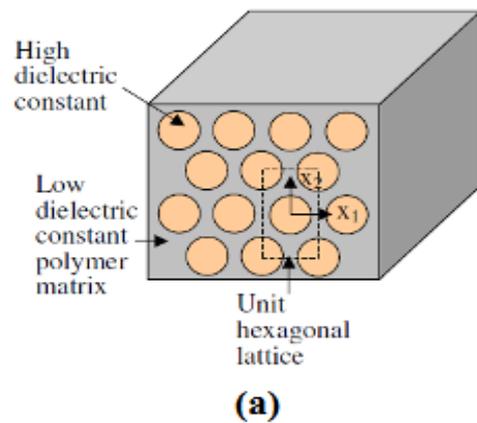


Fig. 3(a) Polymer composite with hexagonal arrangement of high dielectric constant fiber inclusions. Ratio of dielectric constant of fibers to that of the matrix is taken to be 100. In FEM analyses we take $a = 1$. 3(b) Unit Hexagonal lattice cell and applied boundary conditions. ϕ is the electric potential.



Fig. 3(a) and 3(b) the finite element analyses results for a particular case of hexagonal arrangement of fibers with fiber radius being 0.75 times the lattice parameter $a = 1$. To obtain the effective dielectric constant, constant potentials $\phi=1$ and $\phi=-1$ are applied on the left and right faces of the unit cell, respectively (amounting to unit electric field in the horizontal direction), with zero flux boundary conditions on the top and bottom faces and average flux density passing through the left face is evaluated. The effective dielectric constant is then evaluated to be equal to the ratio of average electric flux density passing through the left face to the electric permittivity of vacuum. Or in other words, effective in plane permittivity would be equal to the average flux density passing through the left face. While Fig. 3(a) and 3(b) shows the hexagonal arrangement of fibers, unit cell structure, and applied boundary conditions, Fig. 3(c), Fig. 3(d), and Fig. 3(e) show the electric potential and electric flux density profiles for the applied boundary conditions. It is evident that the electric flux density for most part is routed through the high dielectric constant fibers. Fig. 3(f) makes this observation more apparent. Effectivedielectric constant is evaluated by taking the average of electric flux density plotted in Fig. 3f.

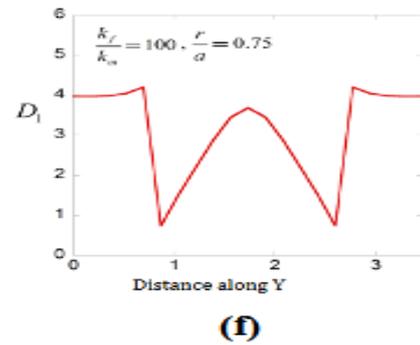
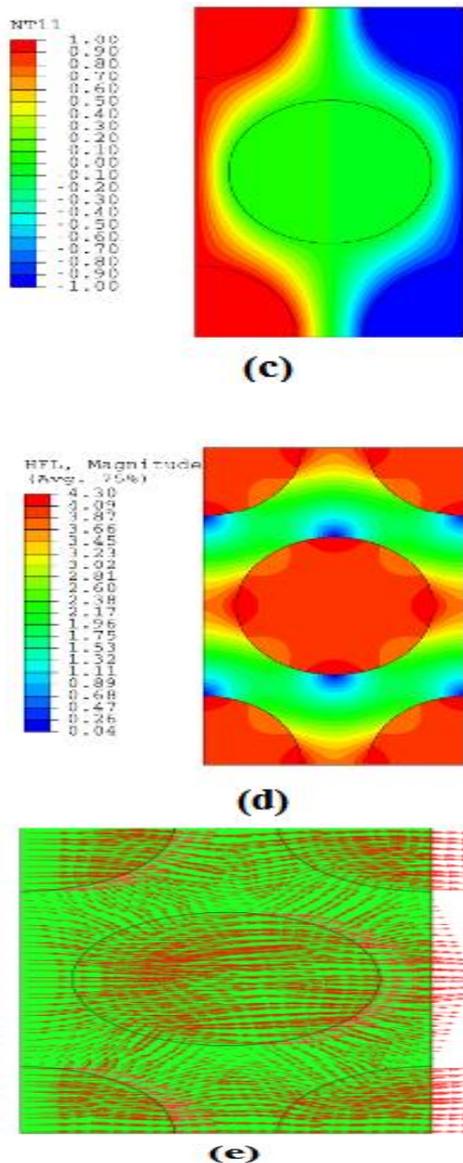
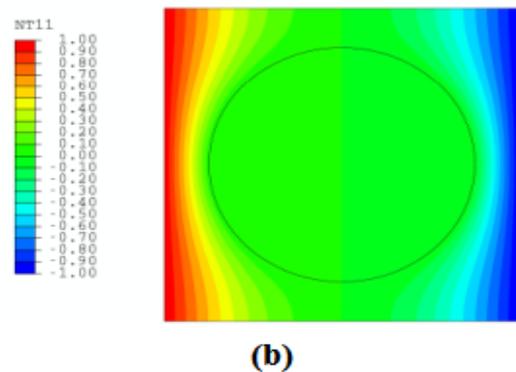
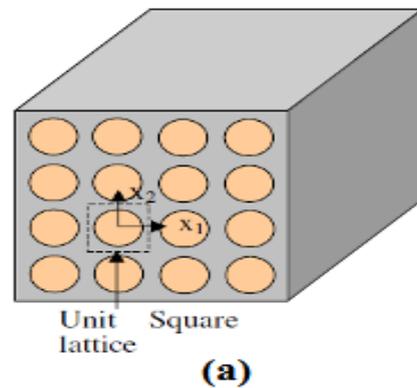


Fig. 3(c) Electric potential profile across the unit lattice for an average electric field of unity along the horizontal direction. 3(d) Resultant electric flux density profile. 3(e) Electric flux density flow patterns. 3(f) Electric flux density along the vertical direction on the left face of the unit lattice. Its average value would be identical to effective in plane direction constant.

Fig. 4(a), Fig. 4(b), Fig. 4(c), Fig. 4(d) and Fig. 4(e) shows similar results as plotted in Fig. 3 but for square arrangement of fibers. It can be seen that the electric potential and electric flux density profiles are very different in both the cases. Nevertheless, even in this case, it can be seen that the electric flux is mainly route through the fibers.

Fig. 5 summarizes the results for periodic (square and hexagonal arrangements) and random microstructures. Results are shown for both in-plane as well as out-of-plane effective dielectric constants as a function of volume fraction of fibers. Note that the out-of-plane dielectric constant does not vary with the type of microstructure as the fields within the fibers and polymer matrix are decoupled for electric fields applied along the fiber direction.



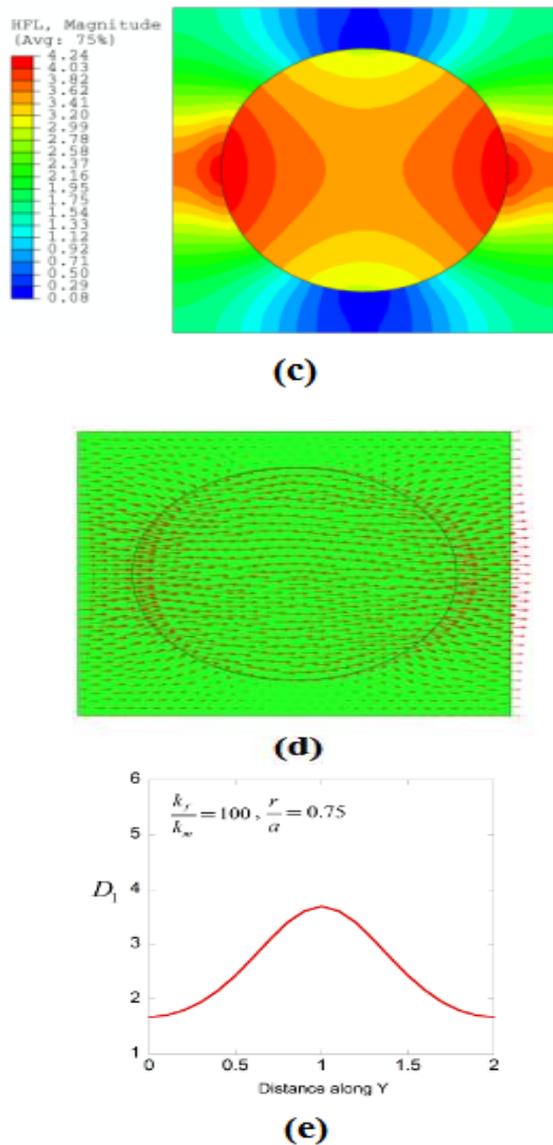


Fig. 4(a) Polymer composite with square arrangement of high dielectric constant fiber inclusions. Ratio of dielectric constant of fibers to that of the matrix is taken to be 100. In FEM analyses we take $a = 1$. 4(b) Electric potential profile across the unit lattice for an average electric field of unity along the horizontal direction. 4(c) resultant electric flux density profiles. 4(d) Electric flux density flow patterns. 4(e) Electric flux density along the vertical direction on the left face of the unit lattice. Its average value would be identical to effective in plane direction constant.

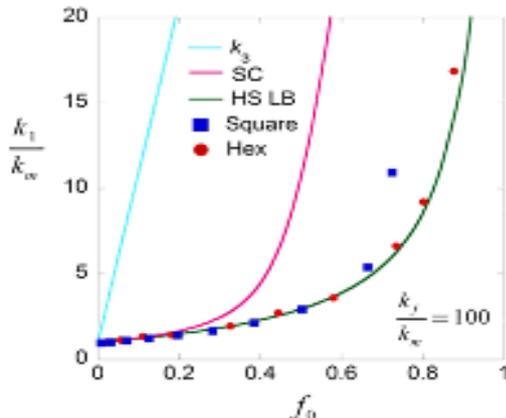


Fig. 5 Effective in-plane and out-of-plane dielectric constants k_1 and k_3 as a function of volume fraction for periodic microstructures with square and hexagonal lattice arrangements and Hashin Shtrikhman lower bound (HS LB) and self-consistent (SC) estimates for random composites.

VI. CONCLUSION

Effective dielectric constant of polymer composites is significantly higher than the dielectric constant for the matrix polymer only at percolation volume fraction if charge injection effects are absent or low. Nano composites can have colossal dielectric constants if significant charge injection effect is present. Only one way to increase the dielectric constant of the Electroactive polymer is by filling the high charge conducting filler particles.

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APPENDIX

Table 1: Maximum performance of various actuator materials at room temperature

Actuator Material (Specific type)	Strain (%)	Pressure (MPa)	Young's Modulus (MPa)	Energy Density (J/g)	Coupling Efficiency k ₂ (%)	Electric Field (V/μm)	Dielectric Constant (@ 1 KHz)
Piezoelectric Ceramic PZT	0.2	110	550	0.013	52	4	1500
Piezoelectric <001> oriented PZN-8%PT single Crystal	1.7	131	7700	0.13	81	12	5000 ^c
Polyurethane (Deerfield PT6100S)	11	1.6	11	0.087	21	160	7
Silicone (Nusil CF19-2186)	32	1.36	1	0.22	54	235	2.8
Silicone (Dow corning HS3)	41	0.13	0.125	0.026	65	72	2.8
Piezoelectric P(VDF-TrFE) with 68:32 mol-%	0.2	6.6	3300	0.0035	8	150	20
Irradiated P(VDF-TrFE) with 68:32 mol-%	5	20	400	0.27	9	160	50
P(VDF-TrFE- CFE) with 65:35:4 mol-%	4.5	50	1100	0.59	31	120	55
P(VDF-TrFE- CFE)/PANI	1.5	8	535	0.032	n.a	9.5	>1000
Natural Muscle (Human skeletal)	>40	0.35	1	0.07	n.a	n.a	n.a
PU-CuPc nano- composite with 3.5 Vol% CuPc	8	1.6	80	0.11	n.a	11.5	>1000