Journal of Engineering Mechanics, Volume 145, Issue 12, December 2019
DOI:10.1061/(ASCE)EM.1943-7889.0001674

Piezoelectric Metamaterial with Negative and Zero Poisson's Ratio

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Abstract

This study presents the finite-element based micromechanical modeling approach to obtain the electromechanical properties of the piezoelectric metamaterial based on honeycomb (HC) cellular networks. The symmetry of the periodic structure was employed to derive mixed boundary conditions (MBCs) analogous to PBCs. Three classes of hexagonal HC cellular networks, namely, conventional HC (CHC), a re-entrant HC (RE) and a semi-re-entrant HC (SRE) were considered. The representative volume elements (RVEs) of these three classes of cellular materials were created, and finite element analyses were carried out in order to analyze the effect of orientation of the ligament on their effective electromechanical properties and their suitability in specific engineering applications. The longitudinally poled piezoelectric HC cellular networks showed an enhanced behavior as compared to the monolithic piezoelectric materials. Moreover, longitudinally poled HC cellular networks demonstrated that, as compared to the bulk constituent, their hydrostatic figure of merit increased and their and acoustic impedance decreased by one order of magnitude, respectively, indicating their applicability for the design on hydrophones. Moreover, results showed that cellular metamaterial with tunable electromechanical characteristics and variety of auxetic behaviors such as negative, positive or zero Poisson's ratios could be

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developed. Such novel HC network-based functional cellular materials likely to facilitate the design of light-weight devices for various next-generation sensors and actuators.

Keywords: cellular materials; effective electromechanical response, honeycomb cellular networks, micromechanical modeling, piezoelectric materials, smart auxetic structures.

Introduction

Piezoelectric materials (PMs) play a key role in advanced multifunctional composites industry by virtue of their unique electromechanical coupling characteristics (Muliana 2011). PMs have found applications in actuators, sensors, ultrasound imagers, hydrophones and echo-cardiogram (Marselli et al. 1999). In these devices, PMs convert mechanical energy into electrical or vice-versa. For example, in sensing applications, PMs require high sensitivity and low acoustic impedance (Alkhader et al. 2015; Lethiecq et al. 2004). Existing bulk piezoelectric polymers show low acoustic impedance and sensitivity while ceramics type piezoelectric materials have high acoustic impedance and sensitivity (Kar-Gupta and Venkatesh 2006). Several studies were conducted to enrich the coupling characteristics between the mechanical and electrical properties in piezoelectric materials by either embedding constituents (Topolov and Bowen 2008) or by introducing the porosity (Hikita et al. 1983).

Piezoelectric composites were tailored to exhibit improved piezoelectric activity and mechanical flexibility (for example, active fiber composites, AFM) (Elhajjar et al. 2013). However, the porosity can significantly enhance the performances of medical diagnostic devices (Smith 1989) and hydrophones (Geis et al. 2000). Both piezoelectric composites (e.g., (Skinner et al. 1978); (Ramesh et al. 2006); (Richard et al. 2004))., and porous PMs (e.g., Nagata et al. 1980; Hikita et al. 1983; Ting 1985; Bast and Wersing 1989; Bowen et al. 2004; Piazza et al. 2005; Zhang et al. 2007; Lee et al. 2007) showed potential in obtaining low acoustic impedance and high piezoelectric sensitivity. However, currently, piezoelectric composites demonstrate properties far from the ideal electromechanical characteristics.

In designing the porous PMs and the piezoelectric composites, the spatial distribution of the two phases controls the effective enhancement of the electromechanical properties (Levassort et al. 1998). Therefore, the concept of phase connectivity was defined (Newnham et al. 1978). To understand the role of connectivity of porosity on the properties of PMs, several experimental studies were conducted by considering numerous configurations of porosity, such as, embedded porosity in a PMs (3-0 foam) (Marselli et al. 1999; Kara et al. 2003; Ueda et al. 2010); long cylinder-like porosity (3-1 foam) (Wirges et al. 2007) and open-cell like porosity (3-3 foam) (Nagata et al. 1980). Results demonstrated that porosity help to enhance the sensitivity of PMs.

Several analytical studies were conducted to estimate the electromechanical characteristics of porous PMs by considering simplified configurations of porosity (such as 3-0 and 3-1) (Banno 1985, Dunn and Taya 1993b, Mikata 2001, Bowen and Topolov 2003). Several micromechanical models based finite element frameworks were developed for predicting electromechanical properties of 3-0, 3-1 and 3-3 type porous PMs and hence addressed more complex microstructure (Kar-Gupta and Venkatesh 2008, Bosse et al. 2012). These studies established that behavior of porous PMs governed by the porosity level, pore shape, pore direction, and their distribution, cellular interconnectivity and the direction of the poling (Iyer et al. 2015).

Piezoelectric cellular materials (e.g., Challagulla and Venkatesh 2012, Bauer et al. 2014) and piezoelectric architectured foams (Fang et al. 2007; Ueda et al. 2010) are the possible subclasses of the porous PMs. These subclasses can be used to tailor the microstructure for developing novel materials with the desired multifunctionality for specific applications (Wadley 2006).

Among Piezoelectric cellular solids and architectured foams, the 3-1 type cellular honeycomb (HC) configuration is highly in use due to its simplicity, utility, and workability with structures. Based on the ligament orientations the HC architecture may produce deformation with positive (conventional), negative (auxetic) and zero Poisson ratio's (Grima et al. 2010). Auxetic materials are attractive due to their counterintuitive response under deformation and improved properties. The structural behavior of HC cellular networks with passive elastic anisotropic ligaments representing auxetic effects has been extensively studied (Grima et al. 2010, Masters and Evans 1996, Gibson and Ashby 1997, Zhu et al. 1997). These studies established that the ligament orientation of cellular network governed the hexagonal HC architecture-property relationship (Alkhader and Vural 2009; Papka and Kyriakides 1999).

In the case of the PMs with cellular networks, there is an auxiliary complexity in finding out optimized architecture-property relationship due to the electromechanical coupling parameters. The dielectric, elastic, and electromechanical coupling anisotropy of the ligament base material, the orientation of the poling direction are the main features of the mentioned problem. There is no analytical model available in the literature that could yield the electromechanical properties of the piezoelectric HC cellular network. Moreover, the relationship between the overall electromechanical properties and the microstructure features for the complete family of piezoelectric HC cellular networks is still not available. Recent experimental work on the fabricated 3-1 type auxetic lattice structure from bulk PZT piezo-ceramic showed promising electromechanical properties with negative in-plane Poisson's ratio of −2.05 (Fey et al. 2016). These experimental findings motivate the present study.

In this paper, micromechanical modeling based FE computational framework is proposed to characterize the effects of poling direction, anisotropic material behavior and ligament orientation on the electromechanical properties of piezoelectric hexagonal HC cellular solids.

Architecture of Piezoelectric Cellular Material

We have considered three types of HC piezoelectric architected materials as shown in Figure 1. These included conventional hexagonal HC structure (CHC), a re-entrant HC (RE) (which generates auxetic behavior) and a semi-re-entrant HC (SRE). In specific configurations, the proposed HC cellular networks can yield a variety of auxetic behavior, i.e., positive, negative and zero Poisson's ratio.

Figure 1 shows the proposed architectures and the four parameters that are used to describe the geometry of HC cellular network. The parameters h, l, t, and θ are referred to as the height of vertical rib, the angular rib length, the rib thickness, and the rib angle, respectively, as shown in the unit cell (UC) of each HC network in Figure 1. We fixed the h=10mm, l=4mm, t=1mm. To obtain different auxetic behaviors, the orientations of the ligaments (θ) of the cellular network were varied as shown in the corresponding unit cell (UC). The HC cellular network ligament material is made of soft PM (i.e., PZT-5H). The HC cellular networks were assumed to be poled in two directions, i.e., aligned with the pore axis (longitudinally poled (LP) network) and perpendicular to the pore axis (transversely poled (TP) network).

Constitutive Model of Piezoelectric Cellular Material

The linearized constitutive equations for a piezoelectric material are given by:

$$\begin{aligned} \varepsilon_{ij} &= S^{E}_{ijkl}\sigma_{kl} + d_{kij}E_k \\ D_i &= d_{ikl}\sigma_{kl} + \kappa^{\sigma}_{ij}E_j \end{aligned} \tag{1}$$

The ε_{ij}, σ_{ij}, D_i and E_i represent the strain tensor, stress tensor, electric displacement vector, and electric field vector, respectively. The material constants S_{ijkl}^{E}, κ_{ij}^{σ}, and d_{ijk} referred to as the fourth-order compliance tensor measured at zero or constant electric field, the second order dielectric tensor measures at zero or constant stress and the components of the piezoelectric strain tensor. Further discussions on the PM constitutive relations can be found elsewhere (Yang 2006). To obtain the homogenized electromechanical properties ($\bar{S}_{ijkl}^{E}$, $\bar{d}_{kij}$, and $\bar{\kappa}_{ij}^{\sigma}$) of the UC, Eq. (1) can be written in terms of average stresses $\bar{\sigma}_{ij}$, strains $\bar{\varepsilon}_{ij}$, electric field $\bar{E}_i$, and electric displacement $\bar{D}_i$.

$$\begin{aligned}\bar{\varepsilon}_{ij} &= \bar{S}_{ijkl}^{E}\bar{\sigma}_{kl} + \bar{d}_{kij}^{E}\bar{E}_k \\ \bar{D}_i &= \bar{d}_{ikl}^{\sigma}\bar{\sigma}_{kl} + \bar{\kappa}_{ij}^{\sigma}\bar{E}_j\end{aligned} \quad (2)$$

According to Eq. (2), the modeling of electromechanical behavior of the cellular PMs requires computing 45 independent material constants, comprising 6 dielectric, 18 piezoelectric and 21 elastic constants.

We considered the linearized electromechanical coupled constitutive relations for each ligament material to obtain the homogenized electromechanical properties of the proposed cellular network. These linearized constitutive relations are simple and, arguably, the most practical method for describing the electromechanical behavior of cellular network at the macroscale (Challagulla and Venkatesh 2013, Iyer et al. 2014, Kar-Gupta and Venkatesh 2006, Sigmund et al. 1998). These assumptions lead to an orthotropic linear electromechanical material model. However, there are advanced models available in the literature (Misra and Poorsolhjouy 2016, Rosi and Auffray 2016, Dell'Isola et al. 2018) that showed that linear microelements in an RVE would produce a nonlinear homogenized response. The geometry-driven non-linearity prevents the use of linear models as linear models do not correctly exhibit the complexity and inter-connectivity of different parameters involved in formulating the problem. In the present study, we do not consider models that account for geometry-driven nonlinearity. However, it will be interesting to analyze the proposed cellular network using such advanced models that account for higher order gradient theory and geometry-driven non-linearity.

It should be emphasized here that the use of linearized electromechanical coupled constitutive relations has some implications. For example, higher order gradient continuum models have been recently used by several researchers to investigate the dispersive wave propagation in strain gradient elastic media. Contrary to classical elasticity within the strain gradient framework, Rosi and Auffray (Rosi and Auffray 2016) showed that the wave propagation in hexagonal lattices becomes anisotropic and group velocity was proven to be different from energy velocity and should be treated as different quantities. Misra and Poorsolhjouy (Misra and Poorsolhjouy 2016) derived a micro-morphic continuum model for the elasticity of granular media, and the dispersion graphs have presented showing the relationship between the dispersion behavior and the micro-scale parameters. Contrary to classical elasticity, where all the waves will be of an acoustic type, and there will be no possibility of frequency band gaps, the micro-morphic continuum model can show band gaps over a large range of wave numbers. Their results indicate that materials with specific wave propagation behaviors can be designed that can replace existing PMs which are commonly used in damage identification or vibration control applications.

Micromechanical Finite Element Model for Piezoelectric Cellular Materials

Recently, Khan and Abu Al-Rub (Khan and Abu Al-Rub 2017, 2018a; b) developed an FE-based UC homogenization method to predict the overall mechanical properties of periodic architectured materials based on the microstructure geometry and its base material properties. In this study, we have extended the framework to calculate the overall properties of architected and periodic cellular PMs. Figure 1 shows the UCs of the three types of hexagonal HC structures. Periodic boundary conditions (PBCs) are imposed on UC (Luxner et al. 2005, Kanit et al. 2003; Khan and Muliana 2009, Choudhry et al. 2016) that yield the responses of infinitely repeating patterns of architecture (Jiang et al. 2002, Abueidda et al. 2015). PBCs give quite reasonable estimates of the properties as compared to the homogeneous displacement and homogeneous traction boundary conditions (Zohdi and Wriggers 2005, Xia et al. 2003). Using the proposed framework, it is possible to characterize the linear electromechanical response of piezoelectric architecture materials completely.

Finite Element Models

FE models of the HC based cellular materials were created by varying the ligament orientation ranging from 30-60 degree which corresponds to porosity values ranging from 50-85%. The FE analyses on the UC were performed with the ABAQUS/Standard. A soft piezoelectric material (PZT-5H) was considered as a base material for the cellular material and its electromechanical properties are presented in Table 1. A representative FE model of HC RE is shown in **Figure** 2 with its corresponding 6 boundary faces direction notations. UC has meshed with 10-noded quadratic piezo-electric tetrahedron elements (C3D10E). Each node in C3D10E has a total of four degrees of freedom (DOF), three displacements (u_1, u_2, u_3), and one electrical potential (ϕ). To avoid the rigid body motion of the UC, under electrical loading cases, the locations of arbitrary points A, B, and C that are constrained specifically are also shown in **Figure** 2.

The cellular materials were assumed to be poled longitudinally and transversely. We have assumed that every region of the FE model considered in this study was poled uniformly in one preferred direction. Uniform poling of the piezoelectric HCs normal to transverse porosity direction is a very challenging task. However, fabrication of the cellular HCs porous networks with uniform poling along the longitudinal direction has been successfully demonstrated (Fey et al. 2016).

Boundary conditions and homogenization for RVE

PBCs are usually applied to the UC to ensure that UC represents the response of the whole architected foam. Moreover, the displacements compatibility and the electric potential continuity across neighboring UC boundaries are assured (Iyer and Venkatesh 2010, Iyer and Venkatesh 2011).

Xia et al. (Xia et al. 2003) developed periodic boundary conditions for a UC in terms of average contractions and stretches ($c_i^j, i = j = 1,2,3$)and shear deformations ($c_i^j, i \neq j$) of the UC model,

$$u_i^{j+}(\mathrm{x,y,z}) - u_i^{j-}(\mathrm{x,y,z}) = c_i^j \qquad (i, j = 1,2,3) \tag{3}$$

The $u_i^{j+}(\mathrm{x,y,z})$ and $u_i^{j-}(\mathrm{x,y,z})$ are the displacements on the positive and negative X_j directions, respectively. Similarly, the PBCs for the electric potential are given by as follows

$$\phi^{j+}(\mathrm{x,y,z}) - \phi^{j-}(\mathrm{x,y,z}) = \bar{E}_i\,(\mathrm{x}_i^{j+} - x_i^{j-}) \qquad (i = 1,2,3) \tag{4}$$

Where, $\bar{E}_i$ is the applied macroscopic electric field. The Eq. (3) and (4) are sufficient to ensure the displacements compatibility and the electric potential continuity.

One of the requirements of the Eq. (3) is that on the two opposite boundary surfaces, the difference of the displacements of the corresponding points should be specified. However, Li (Li 2008) derived an explicit displacement BCs of UCs representing microstructure of periodic structure taken from the symmetry existent within the structure. In this study, the Li (Li 2008) work has been extended to account for electric charge continuity across neighboring UCs and the mixed boundary conditions are proposed to compute the overall properties of piezoelectric architected materials. Table 2 shows the list of the B.C.s.

To couple micro-macro scale behavior, a homogenization method was adopted to obtain the overall properties of the UC under various global loading conditions. The volume averaging approach was used to compute the average stress and strain as follows:

$$\bar{\sigma}_{ij} = \frac{1}{V}\int_V \sigma_{ij}\left(x,y,z\right)dV \qquad \bar{\varepsilon}_{ij} = \frac{1}{V}\int_V \varepsilon_{ij}\left(x,y,z\right)dV \tag{5}$$

Analogously the average electric fields and electric displacements are defined by

$$\bar{E}_i = \frac{1}{V}\int_V E_i\left(x,y,z\right)dV \qquad \bar{D}_i = \frac{1}{V}\int_V D_i\left(x,y,z\right)dV \tag{6}$$

Considering the traction continuity, the average stress can be written as

$$\bar{\sigma}_{ij} = \frac{R_{ij}}{A_j}\left(\text{no summation on j}\right) \tag{7}$$

Using an electric charge continuity, the macroscopic electric displacement is given by

$$\bar{D}_i = \frac{q_i}{A_i} \tag{8}$$

Eq. (7) shows that the macroscopic stress over the UC can be computed from the total tractions (R_{ij}) and the corresponding surfaces areas (A_j). Similarly, Eq. 8 establishes that the macroscopic electrical displacement over the UC can be computed from the total charge (q_i) and the areas (A_i) of the corresponding boundary surfaces.

Sanity Check

In the literature, there is yet no analytical formulation available that offer explicit computation of the properties of the piezoelectric HC networks while their ligaments following orthotropic material behavior. Hence, the mixed boundary conditions given in Table 2 was applied to one element and 100x100x100 elements cube to evaluate the correctness and appropriateness of the proposed methods. It was found that the proposed approach yields the homogenous properties of the PZT-5H given in Table 1.

Results and Discussion

To compute the effective properties of each unit cells, we applied the appropriate boundary conditions given in **Table** 2. For each load-case, it was ensured that the applied boundary conditions produced only one non-zero component of the macroscopic stress or electric field vector in Eq. (2). Using all set of the B.C.s, the 45 independent material constants could be determined. **Table** 2 shows some of the relations for the calculation of typical material parameters.

The effective properties of each of the three piezoelectric cellular HC network, such as, components of the elastic compliance, $\bar{S}^{E}_{ijkl}$, piezoelectric strain tensors, $(\bar{d}^{E}_{kij}, \bar{d}^{\sigma}_{ikl})$, dielectric stress tensor, $\bar{\kappa}^{\sigma}_{ij}$, and the corresponding elastic constants, $\bar{C}^{E}_{ijkl}$, elastic moduli, $(E^{E}_{ij}, \mathrm{G}^{E}_{kk})$, and Poisson's ratio, were obtained using proposed procedure. For both the LP and TP networks, all material parameters were computed for a wide range of the ligaments' orientation. As the level of auxeticity and UC microstructure are controlled by θ, all the results were shown as a function of varying θ. Since some of the material parameters cannot be directly computed from the FE results, so, the most commonly used material constant of the PMs are derived from the following relations.

$$\begin{aligned}&\left[\boldsymbol{C}^{E}\right]=\left[\boldsymbol{S}^{E}\right]^{-1};\ [\boldsymbol{e}]=[\boldsymbol{d}]\left[\boldsymbol{C}^{E}\right];\ \left[\kappa^{\varepsilon}\right]=\left[\kappa^{\sigma}\right]-[\boldsymbol{d}][\boldsymbol{e}]^{T}\\&\left[\boldsymbol{S}^{D}\right]=\left[\boldsymbol{S}^{E}\right]-[\boldsymbol{d}]^{T}[\boldsymbol{g}];\ \left[\boldsymbol{C}^{D}\right]=\left[\boldsymbol{S}^{D}\right]^{-1};\ [\boldsymbol{g}]=[\boldsymbol{h}]\left[\boldsymbol{S}^{D}\right];\ [\boldsymbol{h}]=\left[\kappa^{\varepsilon}\right][\boldsymbol{e}]\end{aligned} \tag{9}$$

Where $\left[\boldsymbol{C}^{D}\right]$ and $\left[\boldsymbol{S}^{D}\right]$ are the components of the stiffness and compliance tensor measured at zero electric displacements, respectively. The $[\boldsymbol{e}]$, $[\boldsymbol{h}]$, $[\boldsymbol{g}]$ are the piezoelectric stress tensor, strain tensor, and voltage tensor, respectively. The dielectric strain tensor $\left[\kappa^{\varepsilon}\right]$ is computed at zero strain.

Effective Elastic Response

The Compliance matrix coefficients $\bar{S}_{ijkl}^{E}$ were used to calculate the stiffness constants $\bar{C}_{ijkl}^{E}$. **Figure** 3 illustrates the stiffness constants of longitudinally and TP HC structures over various angles 30°, 45°, and 60°. Both LP and TP HC cellular materials showed substantial variations in both in-plane and out-of-plane stiffness coefficients values for different θ. The elastic properties were found to be anisotropic and showed different trends and non-intuitive behavior.

The stiffness constants of the LP and TP HC network (except the in-plane stiffness constants, C_{12}) varied nonlinearly with the increase of the θ. The conventional piezoelectric HCs exhibit considerably superior in-plane stiffness constants C_{12}, C_{13} and C_{22} as compared to the re-entrant and semi re-entrant HC foams, whereas the re-entrant HC foams exhibited significantly improved in-plane stiffness constants such as C_{11}.

For all the HC cellular structures considered, the normal stiffness constants in the longitudinal direction (such as C_{33}) are usually larger than the in-plane stiffness coefficients (such as C_{11} or C_{22}). The high stiffness along the longitudinal direction (3 directions) is a result of stretching dominated mode of deformation while the lower stiffnesses along the in-plane directions (1 or 2 directions) show that the bending dominated deformation behavior. For both poling directions, the highest stiffness was observed for re-entrant HC cellular structures.

However, the out-of-plane normal stiffness constant C_{13} showed higher stiffness as compared to the in-plane stiffness constant C_{12}. The in-plane compliant behavior is due to the fact that the in-plane Poisson's ratio of the HC network is about one order of magnitude higher than the out of plane Poisson's ratio. For example, the in- (v_{12}) and out-of- (v_{13}) plane Poisson's ratio of the conventional HC with an angle of ligaments of 30^0 is 0.44 and 0.034, respectively.

The shear stiffness constants involving the in-plane shear stiffness constant, C_{44}, and out-of-plane stiffness constant, C_{66}, varied nonlinearly with the increase of the ligament angle (θ). The nonlinear relation between the relative density and ligament angle might be the cause of the nonlinear stiffness behaviors. However, with the increase of θ the out-of-plane shear stiffness constant in, C_{55}, showed a linear trend (Zhang and Ashby 1992). Under the out-of-plane shear loading condition, the deformation behavior along the 3-directions is stretching dominated that makes the HC cellular network stiffer. On the other hand, the in-plane shear stiffness constant, C_{44}, showed a nonlinear trend with the increase of θ. During in-plane shear loading, the ligaments of the HC network deform under bending along the directions 1 and 2. Among the three HC cellular networks, the semi re-entrant HC was observed to have highest in-plane (i.e., C_{44}) and out-of-plane shear stiffness's (i.e., C_{55}, C_{66}) both in longitudinally and transversely porous HC cellular materials. In addition, for the proposed HC cellular networks a diverse range of in-plane ν can be obtained (+ve, -ve and zero). **Figure** 5 shows that the biggest positive +ve and -ve in-plane ν's were observed at an angle of 30^0. **Figure** 4 and 5 results demonstrate that the HC based piezoelectric cellular materials can yield unique sets of elastic properties with different level of auxeticity.

Figure 6 shows the displacement and electric potential contour plots for three different load case. For all three HC structures considered, the displacement contours in 1-direction under uniaxial and shear loading loadings are shown in **Figures** 6(a) and (b), respectively. These distributions of displacement fields clearly demonstrated the state of shear and uniaxial deformation behavior. For the loading case-7, the electric field of 0.1MV/m is applied, and the electric potential contours plots are obtained. **Figure** 6(c) shows that for all three HC network, there is a linear variation of electrical potential between opposite faces of loading while a uniform electric potential was observed at the central part of the UCs.

Effective piezoelectric properties

Figure 7 shows the changes in the overall piezoelectric properties as a function of the angle of ligaments for HC cellular networks considered. The poling direction with respect to the porosity axis usually has an insignificant effect on the piezoelectric properties of porous materials (Iyer et al. 2015). However, it is observed for both TP and LP HC networks that the architecture of the HC cellular network does affect some of the piezoelectric properties. The level of magnitude of piezoelectric properties of TP networks are very less as compared to the LP HC network, but variation response as a function of ligament angle has similar trends.

We analyzed both the normal and shear piezoelectric coefficients. It was found that shear piezoelectric properties of HC cellular network exhibited high piezoelectric sensitivity. However, the shear piezoelectric constants showed considerably different behavior as a function of θ. In the case of LP HC network, the e_{24} (LP) was increased, and e_{15} (LP) was decreased significantly with θ for all HC networks. The largest value of e_{15} (LP) was observed at 30 degrees for a re-entrant network; while for e_{24} (LP) the semi-re-entrant HC network was showing the highest values and increased with the values of θ.

The effective normal piezoelectric properties were found to be a function of ligament angle and HC networks architecture. The normal piezoelectric constant, $e_{33,}$ is usually recognized as the most vital parameter for various applications utilizing monolithic PMs. For LP HC network e_{33} (LP) was amplified with the increase in ligament angle, but there was an insignificant variation for e33(TP). In addition, **Figure** 7 demonstrated that even though the e31 of the ligament base material has negative values, the HC network could display both -ve and +ve values of e_{31} values by varying the angle θ. These results indicated that the piezoelectric HC architecture could show a different crystal symmetry as compared to the ligament base material. The LP HC network produced the highest negative values of e_{31} and e_{32}.

Effective dielectric properties

The effective dielectric constants κ_{ij} were computed for all three HC cellular networks as a function of the θ as presented in **Figure** 8. Both LP and TP HC cellular networks showed noteworthy variations in the κ_{ij} values at various angles. The κ_{ij} values of all piezoelectric HC networks (except $\kappa_{11}(\mathrm{LP})$ and $\kappa_{11}(\mathrm{TP})$ for conventional and semi re-entrant HC) increased and varied nonlinearly as a function of ligaments angle. The decrease in the magnitudes of $\kappa_{11}(\mathrm{LP})$ and $\kappa_{11}(\mathrm{TP})$ for conventional and semi re-entrant HC was related to the scattered path of the electric charges along the x-axis. The highest and lowest values of the κ_{ij} were obtained for re-entrant HC $\kappa_{33}(\mathrm{LP})$ and conventional HC, $\kappa_{11}(\mathrm{TP})$, respectively.

Effective Figure of Merit

Various figures of merit (FOM) are of interest to evaluate the usefulness of PMs in industrial applications, such as an ultrasound imager, hydrophones, and energy harvesters. The combination of fundamental electromechanical coefficients (i.e., elastic, piezoelectric and dielectric coefficients as calculated by the above mentioned FEA) can be used to evaluate numerous industrial FOM. For cellular network FOM, the relevant constants in various applications (i.e., hydrophones) are the hydrostatic strain coefficient (d_h), the hydrostatic FOM ($d_h.\mathrm{g}_h$), the acoustic impedance (Z) and electromechanical thickness model coupling factor (k_t) (Kar-Gupta and Venkatesh 2006, Dunn and Taya 1993b). There are some other FOM which are of importance in several different applications. The relevant FOM parameters are given in Table 3. More details on FOMs can be found elsewhere, (Dunn and Taya 1993b).

To assess how much the proposed HC networks enhanced the electromechanical responses the FOM of the HC network is normalized with the FOM of monolithic PM (PZT-5H) as shown in **Figure** 8. For the TP networks, the results show that all the normalized FOMs did not show any enrichment in electromechanical behavior. However, in the LP networks, some of the normalized FOMs varied strongly and marginally as a function of θ. The normalized d_h showed a very minor effect on the variation of θ while a normalized $d_h.\mathrm{g}_h$ was decreased with the increase in the θ and the highest value was found at 30 degrees. The inverse relation between g_{33} and κ_{33} is the cause of such huge improvement in the $d_h.\mathrm{g}_h$. **Figure** 8 shows that the κ_{ij} decreases with the increase of porosity, and as a result, the g_h was increased. The lowest (better) normalized value of Z was found at 30^0 while the Z value increases with an increase in the θ. The numerical results agreed well with the experimentally observed (Bast and Wersing 1989) trend of decrease in the values of Z with the increase of porosity.

The proposed piezoelectric HC cellular material showed an improved behavior for the LP system as compared to the bulk material. For example, the LP HC networks showed one order of magnitude increase in their hydrostatic FOM. However, the acoustic impedance was decreased by one order of magnitude.

The Z-$d_h.\mathrm{g}_h$ relationship is important to be analyzed while designing the transducer and hydrophones. **Figure** 9 shows the Z-$d_h.\mathrm{g}_h$ quid pro quo, where when the porosity of HC networks increases, the $d_h.\mathrm{g}_h$ showed increasing trend while the Z value decreases. Such a quite unique combination of increased sensitivity and reduced acoustic impedances is nearly impossible to obtain using bulk PMs and piezoelectric composites (Dunn and Taya 1993a). These results indicate that it is possible to design architected porous PM with required electromechanical characteristics. Several researchers have shown that auxetic geometry improves the performance of the piezoelectric composite and porous PMs. For piezoelectric composites, Smith (Smith 1991) showed that the polymer matrix with negative Poisson's ratio enhances their performance. Numerous studies combined topology optimization and finite element methods to design optimum topologies of unit cells to obtain a piezoelectric composite with better behavior. Using the topology optimization method proposed by Bendsøe and Kikuchi (Bendsøe and Kikuchi 1988), the optimum design of 1-3 piezoelectric composites for hydrophone applications were obtained by Sigmund et al (Sigmund et al. 1998). Using the criteria to maximize for d_h and $d_h.\mathrm{g}_h$, the obtained optimal 3D porous architected matrix showed auxetic effects in certain directions like re-entrant type HC cellular material. Moreover, the optimized design with porous architecture design increases the values of d_h and $d_h.\mathrm{g}_h$ over monolithic piezoelectric ceramics by factors of more than 10 and 10,000, respectively. A unit cell based topology optimization approach was proposed by Silva, Kikuchi, and co-workers (Silva et al. 1997, Silva et al. 1998) to find the distribution of inclusion and/or voids phases that enhances piezoelectric electromechanical efficiency. Several 2D and 3D auxetic structures (negative Poisson’s ratio) consisting of piezoelectric polymer and architected cellular materials were proposed.

For the proposed piezoelectric HC cellular networks, a desirable elastic, piezoelectric and dielectric properties can be obtained that is nearly impossible to achieve using bulk PMs and piezoelectric composites. The unique set of electromechanical properties is a function of angle so all the properties cannot be improved at once. It requires some techniques to obtain an optimum combination of properties. For example, using topology optimization Sigmund et al. (Sigmund et al. 1998) showed that the cell design should be optimized such that it gives the required strength and electromechanical properties. The proposed 3-1 type piezoelectric cellular networks are limited in terms of their electromechanical properties and strength. Recently, we have shown that 3-3 type piezoelectric metamaterials can provide a good combination of strength and electromechanical properties (Khan and Khan 2019). The 3-3 type piezoelectric porous metamaterials design improves the values of d_h and $d_h.g_h$ over monolithic piezoelectric ceramics by factors of more than 15 and 12,000, respectively.

Next, we analyzed the behavior of normalized electromechanical coupling factors (k_{31}, k_{32}, k_t) and frequency constants (N_{31}, N_{32}, N_t).**Figures** 10(a-c) showed the normalized magnitudes of the k_{3i} and k_t. The k_{31} decreased while k_{32} increased with the increase in angle θ. The k_{31} and k_{32} were equal for the bulk PZT-5H. For all proposed HC cellular materials, for given angles, the k_{31} values ware seen to decrease more comparative to k_{32} because of the anisotropic nature of microstructure. For both modes, the d_{3i} were nearly equal but the higher values of S_{11}^E than S_{22}^E give rise to more decrease in k_{31} than k_{32}. In contrast, to k_{31} and k_{32}, the normalized kt values were one order higher than the monolithic PZT-5H.

Figures 9(d-f) showed normalized plots of N_1 and N_2. For monolithic material, the frequency constants N_1 and N_2 were equal. For all the proposed HC cellular materials, for given angles, the N_1 values were seen to decrease more comparative to N_2. As compared to monolithic piezoelectric materials, the disparity in N_1 and N_2 values were due to the orthotropic constituent properties and the architecture of the HC cellular materials. Moreover, the reverse trend was observed for N_1 and N_2 with an increase in the porosity. The larger decreased in N_1 than N_2 values are due to the higher values of S_{11}^E than S_{22}^E. The normalized Nt decreased with the increase of porosity as shown in

Figure 10(f). The decreased in N_t with increasing the porosity was linked with the increased in the S_{33}^{E} due to decrease in the transverse damping.

The computational analyses show that the relationship between the architecture of HC cellular network and anisotropic nature of constituent properties leads to architecture-dependent elastic, piezoelectric and dielectric properties that vary significantly from the properties of the monolithic material. Among all three proposed HC networks, the CHC cellular materials showed exceptional electromechanical properties. The FE results confirm that the RE and SRE HC network demonstrate a combination of unique mechanical properties with auxetic effects and exceptional piezoelectric properties which cannot be realized from CHC network. Overall, the FE results endorsed that the cellular networks can be tailored to obtain a unique combination of tunable electromechanical properties as per the needs of various practical applications. The proposed novel RE and SRE HC networks have the capacity to design unique next generation actuators and sensors with negative and zero Poisson's ratio.

Conclusions

This study proposed a finite element base micromechanical modeling framework to estimate the elastic, dielectric and piezoelectric of the HC based cellular PMs. Using the internal symmetry of the periodic structure a mixed boundary conditions analogous to PBCs were proposed. The modeling approach was applied to the UCs of conventional, auxetic and semi re-entrant type HC piezoelectric cellular networks and their electromechanical properties were presented. For the longitudinal poled network, the results showed that the HC network exhibited an exceptional combination of piezoelectric properties, i.e., low impedance and more sensitivity, which could not be obtained by monolithic PMs. However, for the TP network, the electromechanical properties displayed insignificant dependence on the porosity and angle of the ligament. The FE results showed that HC network with tunable electromechanical characteristics coupled with auxetic behavior such as negative or zero Poisson's ratio could be produced. Such novel HC network-based functional cellular materials have the capacity to facilitate the design of light-weight devices for various next-generation sensors and actuators.

ACKNOWLEDEMENTS

The authors would like to thank undergraduate students Abdelrahman Alhammadi, Ali Alneyadi and Saeed Alqaydi from Khalifa University of Science and Technology for generating CAD files of the HC cellular networks.

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Table 1. Electromechanical properties of the PZT-5H (poled in 3-direction)

Properties	PZT-5H
$S_{11}^{E} = S_{22}^{E}$ (pm^2 / N)	16.5
$S_{12}^{E} = S_{21}^{E}$ (pm^2 / N)	-4.78
$S_{13}^{E} = S_{23}^{E}$ (pm^2 / N)	-8.45
S_{33}^{E} (pm^2 / N)	20.7
S_{44}^{E} (pm^2 / N)	42.6
$S_{55}^{E} = S_{66}^{E}$ (pm^2 / N)	43.5
$d_{15} = d_{24}$ (pm/ V)	741
$d_{31} = d_{32}$ (pm/ V)	-274
d_{33} (pm/ V)	593
$\kappa_{11}^{\sigma} / \kappa_0 = \kappa_{22}^{\sigma} / \kappa_0$	3130
$\kappa_{33}^{\sigma} / \kappa_0$	3400
Density (ρ)	7500 kg/m^3
Permittivity of free space (κ_0)	8.85x10^{-12} C/Vm

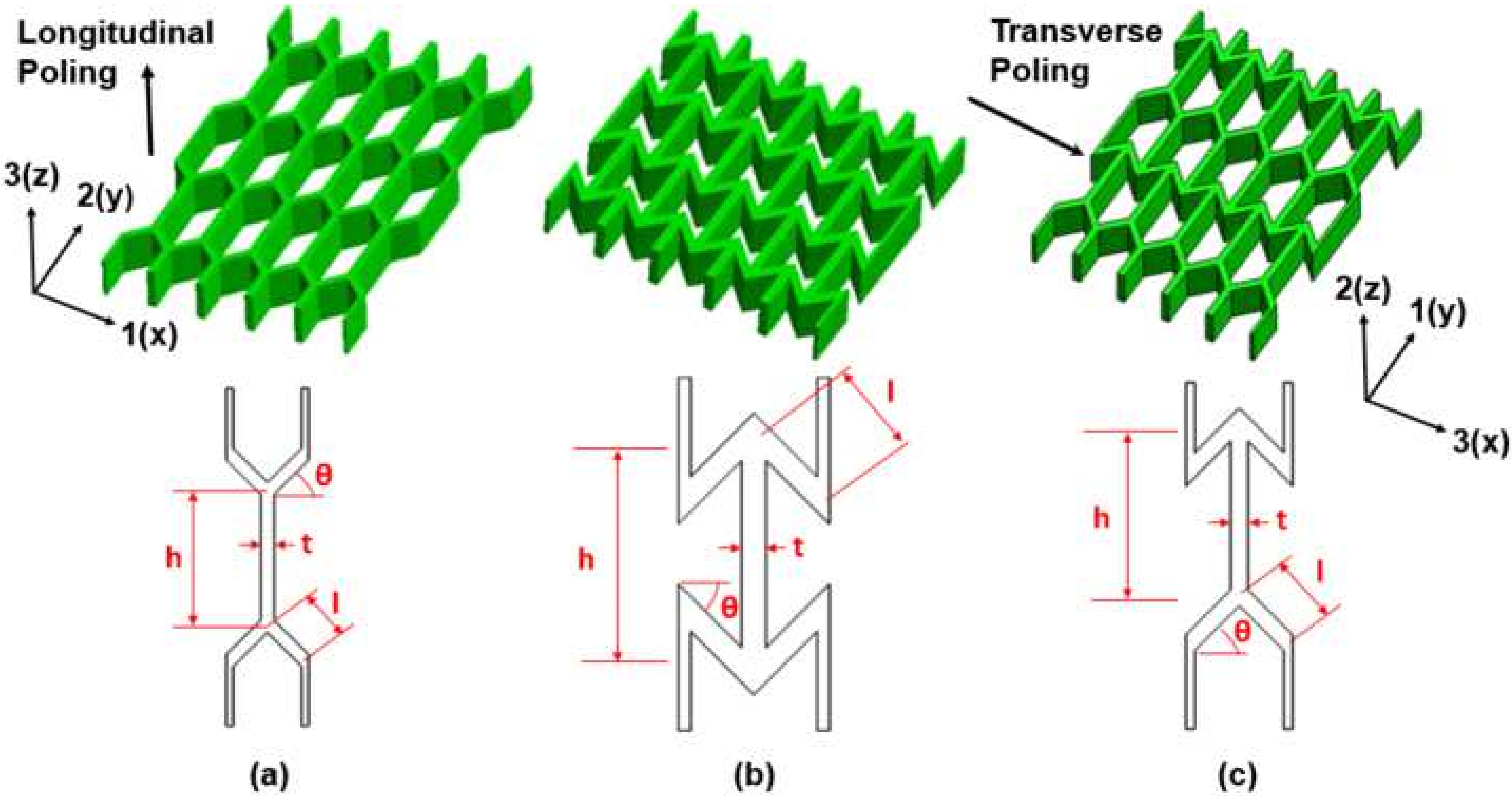
Table 2. Complete set of boundary conditions with computation formulae to determine electromechanical coefficients.

Coefficients	X^-	X^+	Y^-	Y^+	Z^-	Z^+	Relation
	$\bar{u}_i / \bar{\phi}$	$\bar{u}_i / \bar{\phi}$	$\bar{u}_i / \bar{\phi}$	$\bar{u}_i / \bar{\phi}$	$\bar{u}_i / \bar{\phi}$	$\bar{u}_i / \bar{\phi}$	
$\bar{S}^E_{11},\bar{S}^E_{12}$, $\bar{S}^E_{13},\bar{d}^E_{31}$	$0/0$	$\bar{u}_1 / 0$	$0/0$	$-/0$	$0/0$	$-/0$	$\bar{\varepsilon}_{11}/\bar{\sigma}_{11}, \bar{\varepsilon}_{22}/\bar{\sigma}_{11}$, $\bar{\varepsilon}_{33}/\bar{\sigma}_{11}, \bar{D}_3/\bar{\sigma}_{11}$
$\bar{S}^E_{21},\bar{S}^E_{22}$, $\bar{S}^E_{23},\bar{d}^E_{32}$	$0/0$	$-/0$	$0/0$	$\bar{u}_2 / 0$	$0/0$	$-/0$	$\bar{\varepsilon}_{11}/\bar{\sigma}_{22}, \bar{\varepsilon}_{22}/\bar{\sigma}_{22}$, $\bar{\varepsilon}_{33}/\bar{\sigma}_{22}, \bar{D}_3/\bar{\sigma}_{22}$
$\bar{S}^E_{31},\bar{S}^E_{32}$, $\bar{S}^E_{33},\bar{d}^E_{33}$	$0/0$	$-/0$	$0/0$	$-/0$	$0/0$	$\bar{u}_3 / 0$	$\bar{\varepsilon}_{11}/\bar{\sigma}_{33}, \bar{\varepsilon}_{22}/\bar{\sigma}_{33}$, $\bar{\varepsilon}_{33}/\bar{\sigma}_{33}, \bar{D}_3/\bar{\sigma}_{33}$
$\bar{S}^E_{44},\bar{d}^E_{24}$	$\bar{u}_2 = 0,$ $\bar{u}_3 = 0$ /0	$\bar{u}_2 = 0,$ $\bar{u}_3 = 0$ /0	$\bar{u}_1 = 0,$ $\bar{u}_3 = 0$ /0	$\bar{u}_1 \neq 0,$ $\bar{u}_3 = 0$ /0	$\bar{u}_3 = 0$ /0	$\bar{u}_3 = 0$ /0	$\bar{\varepsilon}_{12}/\bar{\sigma}_{12}$, $\bar{D}_2/\bar{\sigma}_{12}$
$\bar{S}^E_{55},\bar{d}^E_{15}$	$\bar{u}_2 = 0,$ $\bar{u}_3 = 0$ /0	$\bar{u}_2 = 0,$ $\bar{u}_3 = 0$ /0	$\bar{u}_2 = 0,$ /0	$\bar{u}_2 = 0,$ /0	$\bar{u}_1 = 0,$ $\bar{u}_2 = 0$ /0	$\bar{u}_1 \neq 0,$ $\bar{u}_2 = 0$ /0	$\bar{\varepsilon}_{13}/\bar{\sigma}_{13}$, $\bar{D}_1/\bar{\sigma}_{13}$
$\bar{S}^E_{66}$	$\bar{u}_1 = 0,$ /0	$\bar{u}_1 = 0,$ /0	$\bar{u}_1 = 0,$ $\bar{u}_3 = 0$ /0	$\bar{u}_1 = 0,$ $\bar{u}_3 = 0$ /0	$\bar{u}_1 = 0,$ $\bar{u}_2 = 0$ /0	$\bar{u}_1 = 0,$ $\bar{u}_2 \neq 0$ /0	$\bar{\varepsilon}_{23}/\bar{\sigma}_{23}$
$\bar{d}^\sigma_{15},\bar{\kappa}^\sigma_{11}$	$*/0$	$-/\bar{\phi}$	$-/-$	$-/-$	$-/-$	$-/-$	$\bar{\varepsilon}_{13}/\bar{E}_1$, $\bar{D}_1/\bar{E}_1$
$\bar{d}^\sigma_{24},\bar{\kappa}^\sigma_{22}$	$-/-$	$-/-$	$*/0$	$-/\bar{\phi}$	$-/-$	$-/-$	$\bar{\varepsilon}_{12}/\bar{E}_2$, $\bar{D}_2/\bar{E}_2$
$\bar{d}^\sigma_{31},\bar{d}^\sigma_{32}$, $\bar{d}^\sigma_{33},\bar{\kappa}^\sigma_{33}$	$-/-$	$-/-$	$-/-$	$-/-$	$*/0$	$-/\bar{\phi}$	$\bar{\varepsilon}_1/\bar{E}_3, \bar{\varepsilon}_2/\bar{E}_3$ $\bar{\varepsilon}_3/\bar{E}_3, \bar{D}_3/\bar{E}_3$

*Points A, B and C are constrained on respective faces (having zero electric potential) to avoid rigid body motion.

Table 3. List of figure of merit.

Parameter	Relation
hydrostatic strain coefficient (d_h)	$d_h = d_{31} + d_{32} + d_{33}$
hydrostatic figure of merit (d_h.g$_h$)	$g_h d_h$ with $g_h = g_{31} + g_{32} + g_{33}$
acoustic impedance (Z)	$Z = \left[\rho C_{33}^{E}\right]^{1/2}$
electromechanical thickness mode coupling factor (k_t)	$k_t = \left[1 - \frac{C_{33}^{E}}{C_{33}^{D}}\right]^{1/2} = \frac{e_{33}}{\left[C_{33}^{D}\kappa_{33}^{\varepsilon}\right]^{1/2}}$
electromechanical coupling factor (k_{3i})	$k_{3i} = \frac{d_{3i}}{\left[S_{ii}^{E}\kappa_{33}^{\sigma}\right]^{1/2}}$; (i=1,2 with no sum on ii)
Frequency constants(N_t)	$N_t = \frac{1}{2}\left[\frac{C_{33}^{E}}{\rho}\right]^{1/2}$
Frequency constants(N_i)	$N_i = \frac{1}{2}\left[\frac{1}{\rho S_{33}^{E}}\right]^{1/2}$; (i=1,2 with no sum on ii)


Longitudinal Poling
3(z)
2(y)
1(x)
Transverse Poling
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(a)
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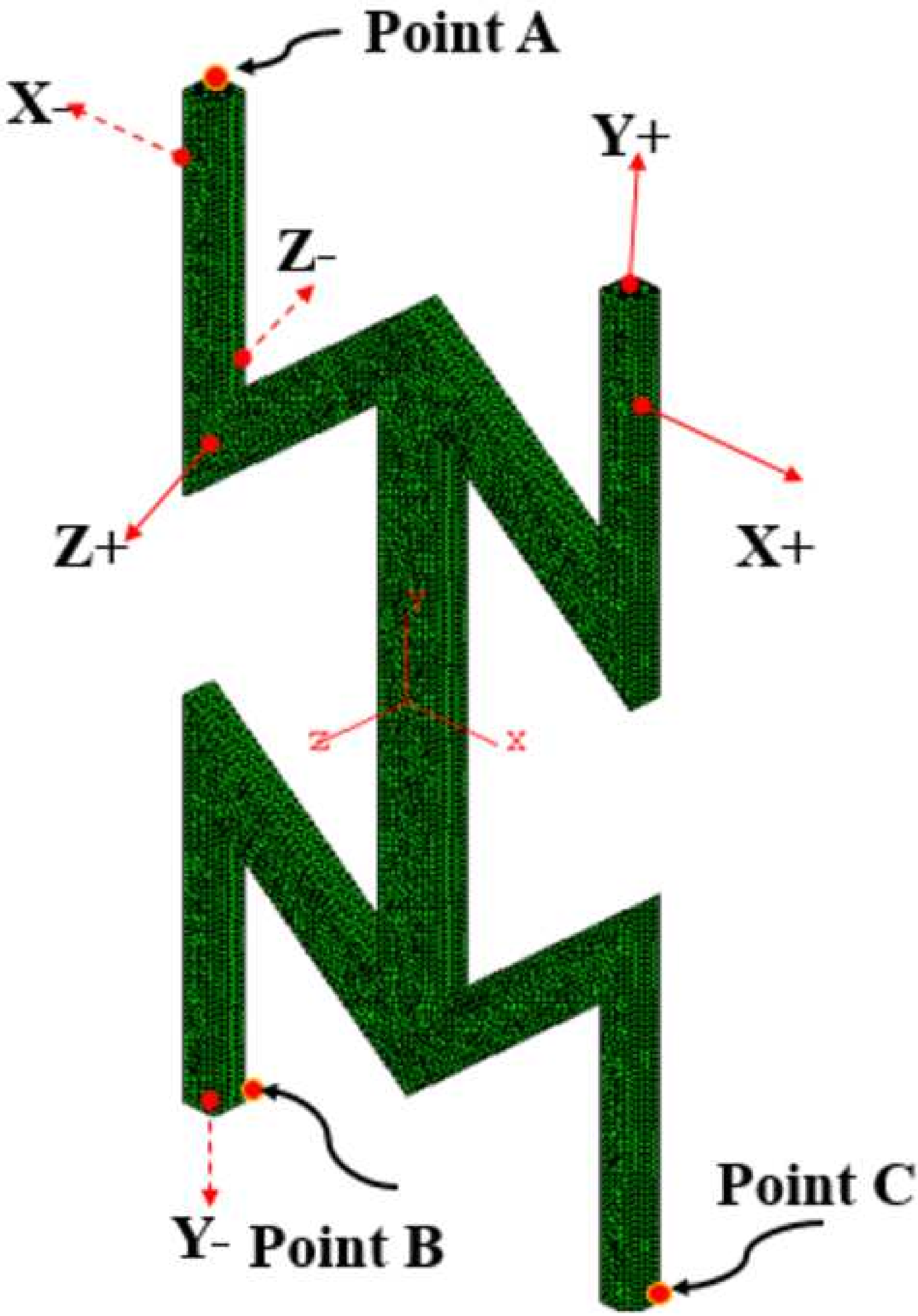
(c)


Point A
X–
Y+
Z–
Z+
X+
y
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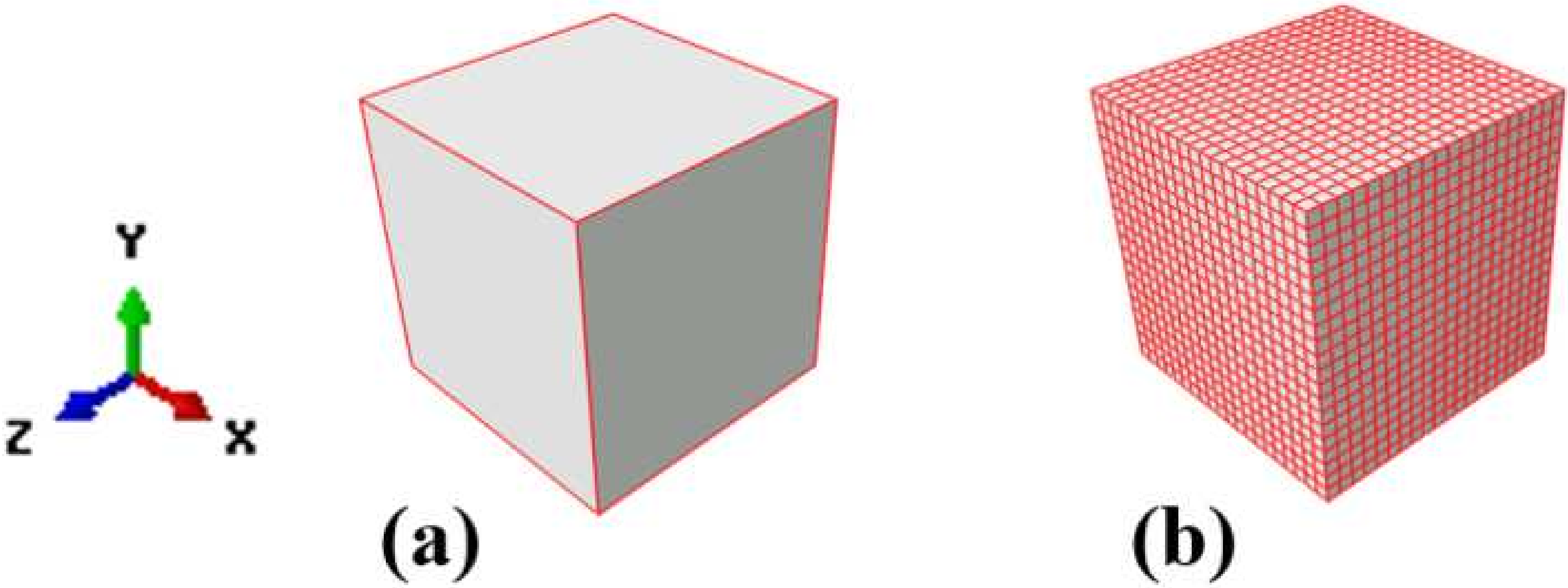
Point C
Y–
Point B

Figure

Click here to access/download;Figure;Figure3.tiff



Figure

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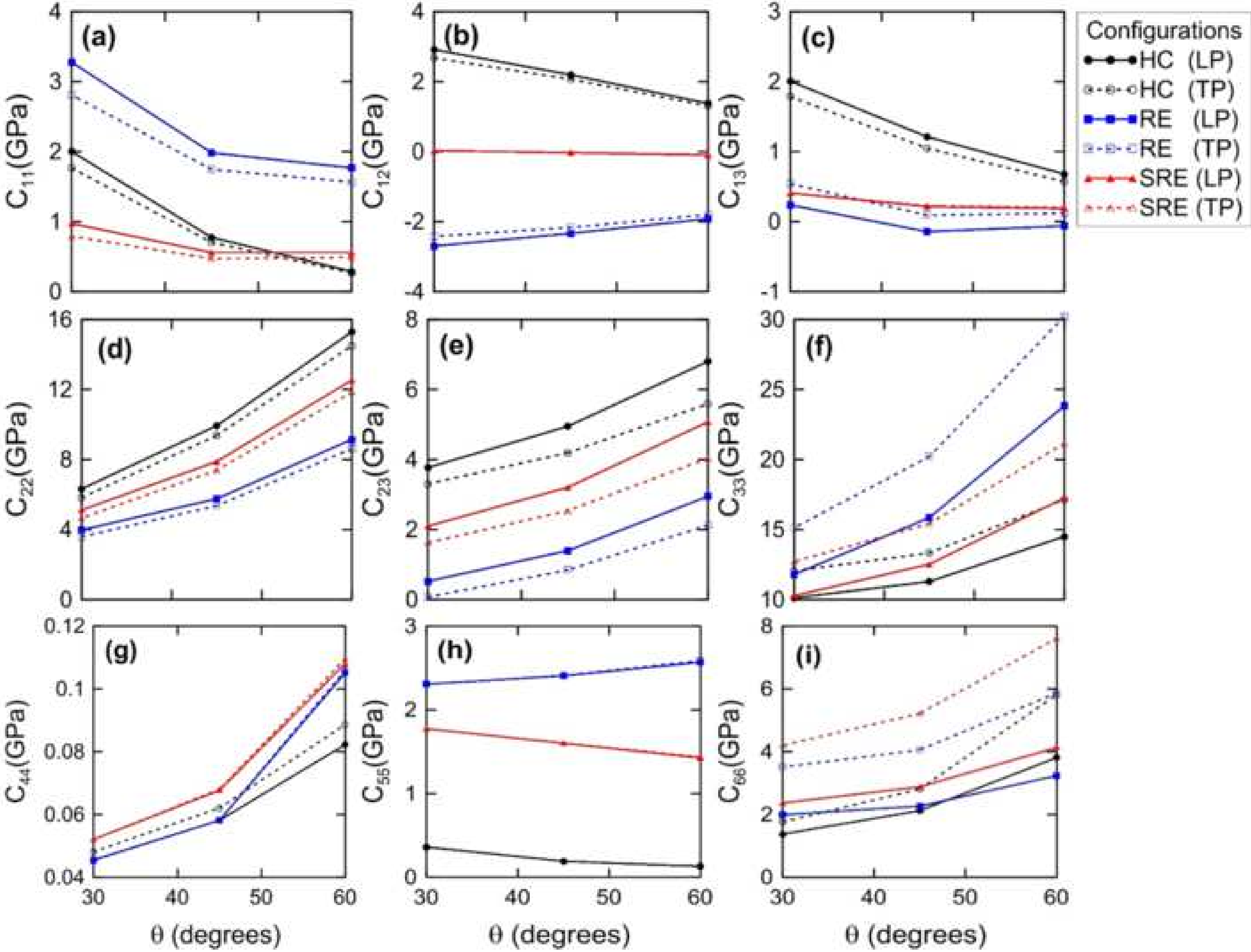
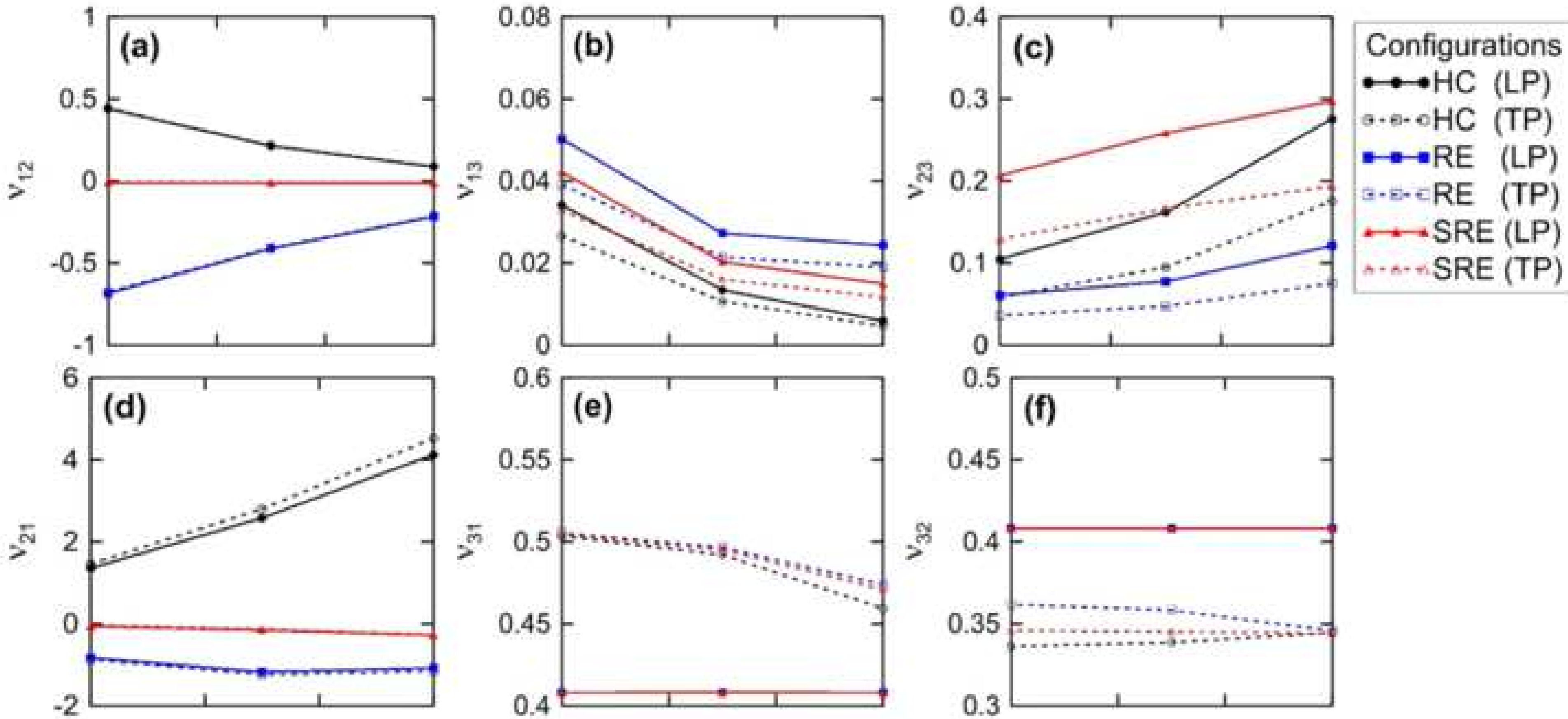
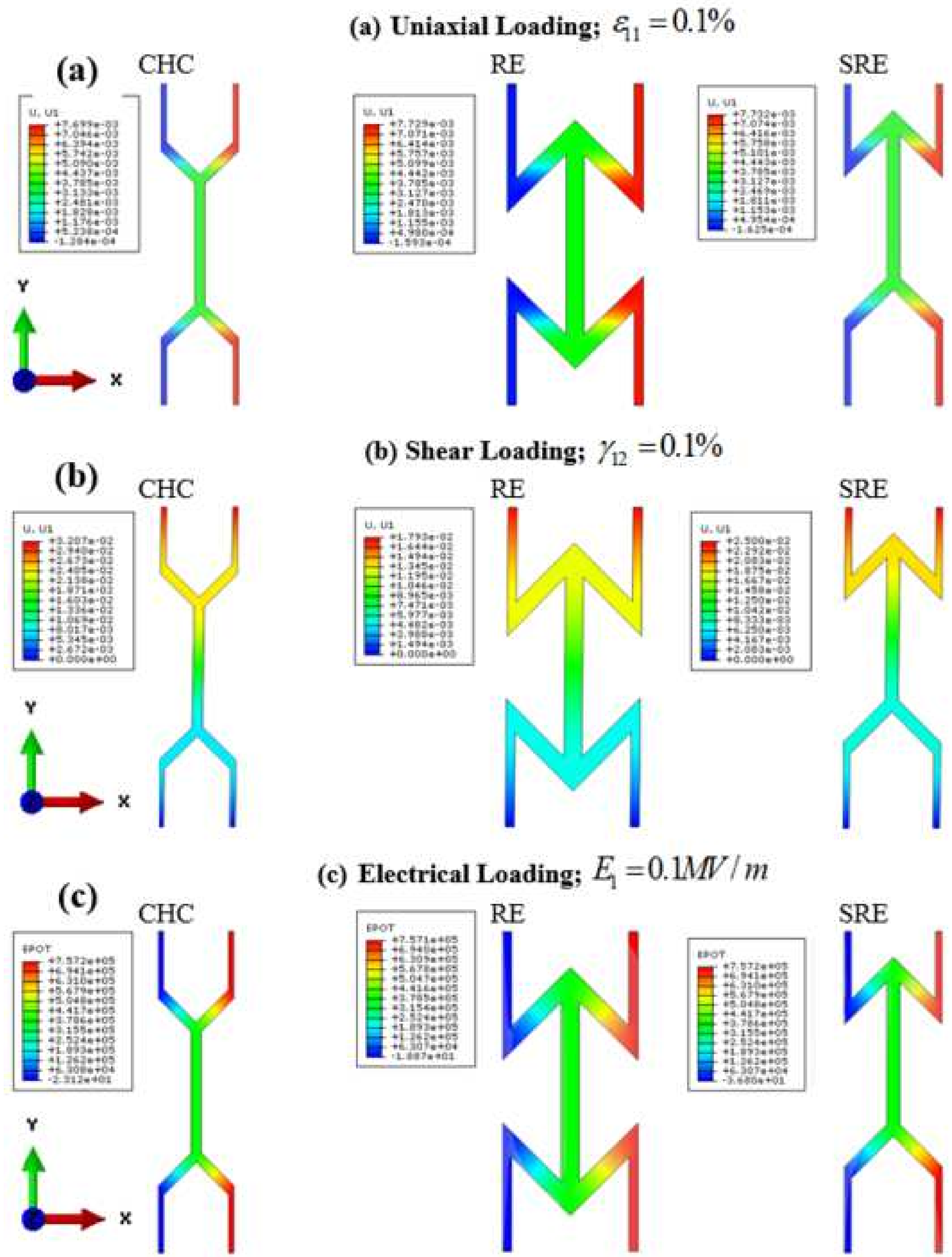
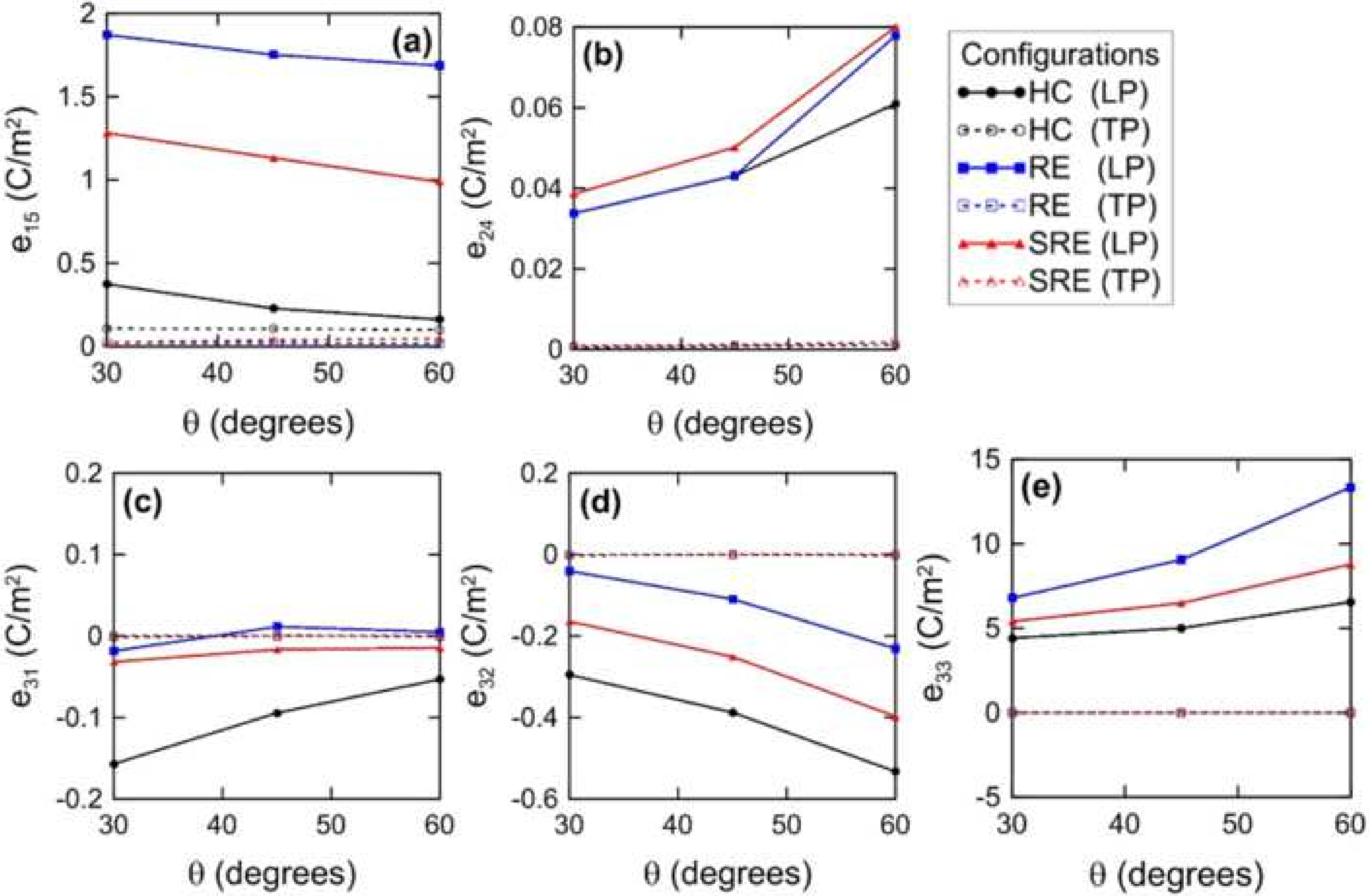



(a)
(b)
(c)
(d)
(e)
(f)
v12
v13
v23
v21
v31
v32
Configurations
HC (LP)
HC (TP)
RE (LP)
RE (TP)
SRE (LP)
SRE (TP)


(a) Uniaxial Loading; $\varepsilon_{11} = 0.1\%$
(a)
CHC
RE
SRE
U, U1
Y
X
(b) Shear Loading; $\gamma_{12} = 0.1\%$
(b)
CHC
RE
SRE
U, U1
Y
X
(c) Electrical Loading; $E_1 = 0.1MV/m$
(c)
CHC
RE
SRE
EPOT
Y
X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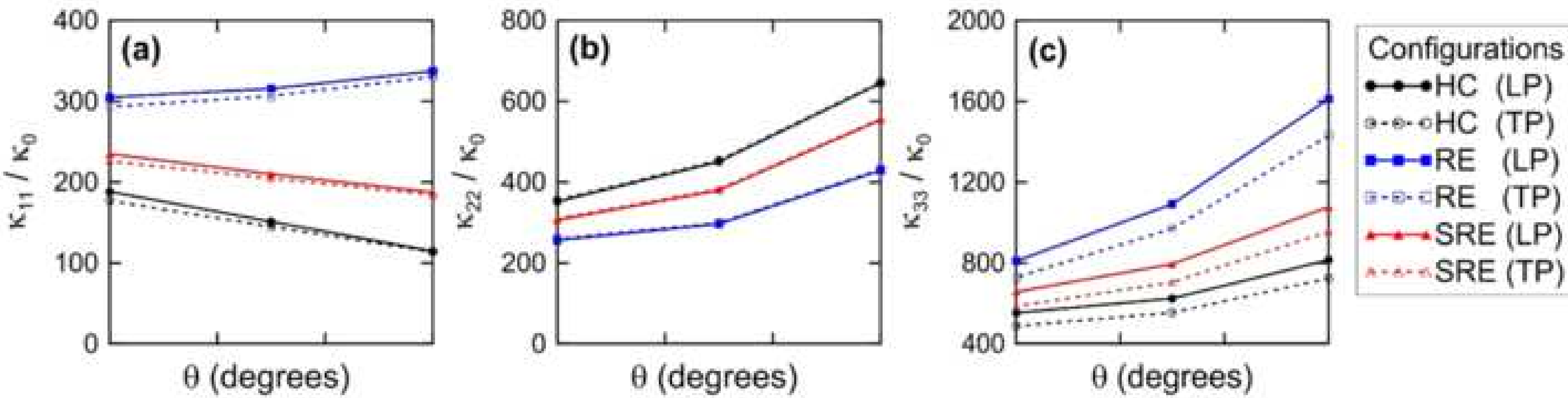
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Figure

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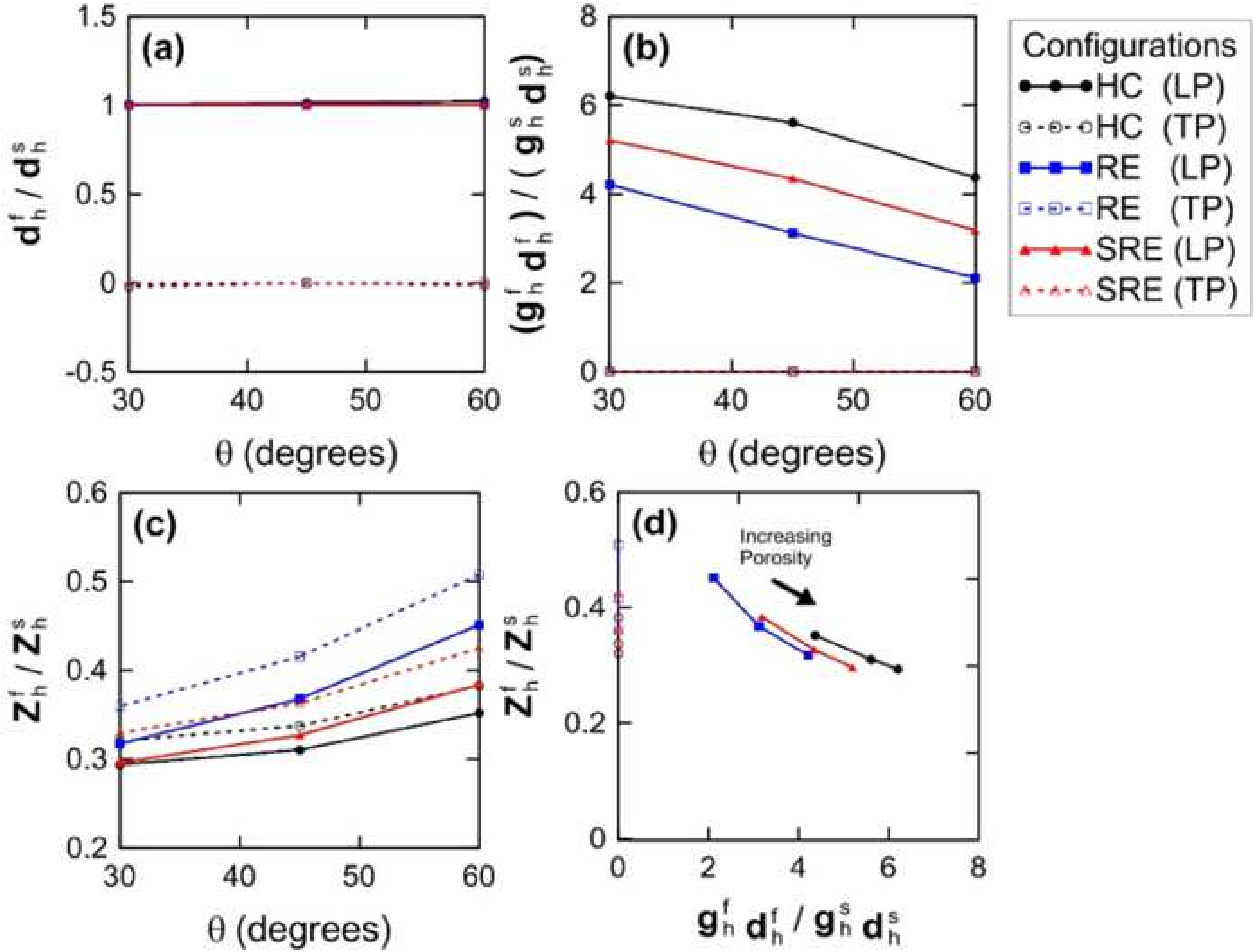

Figure

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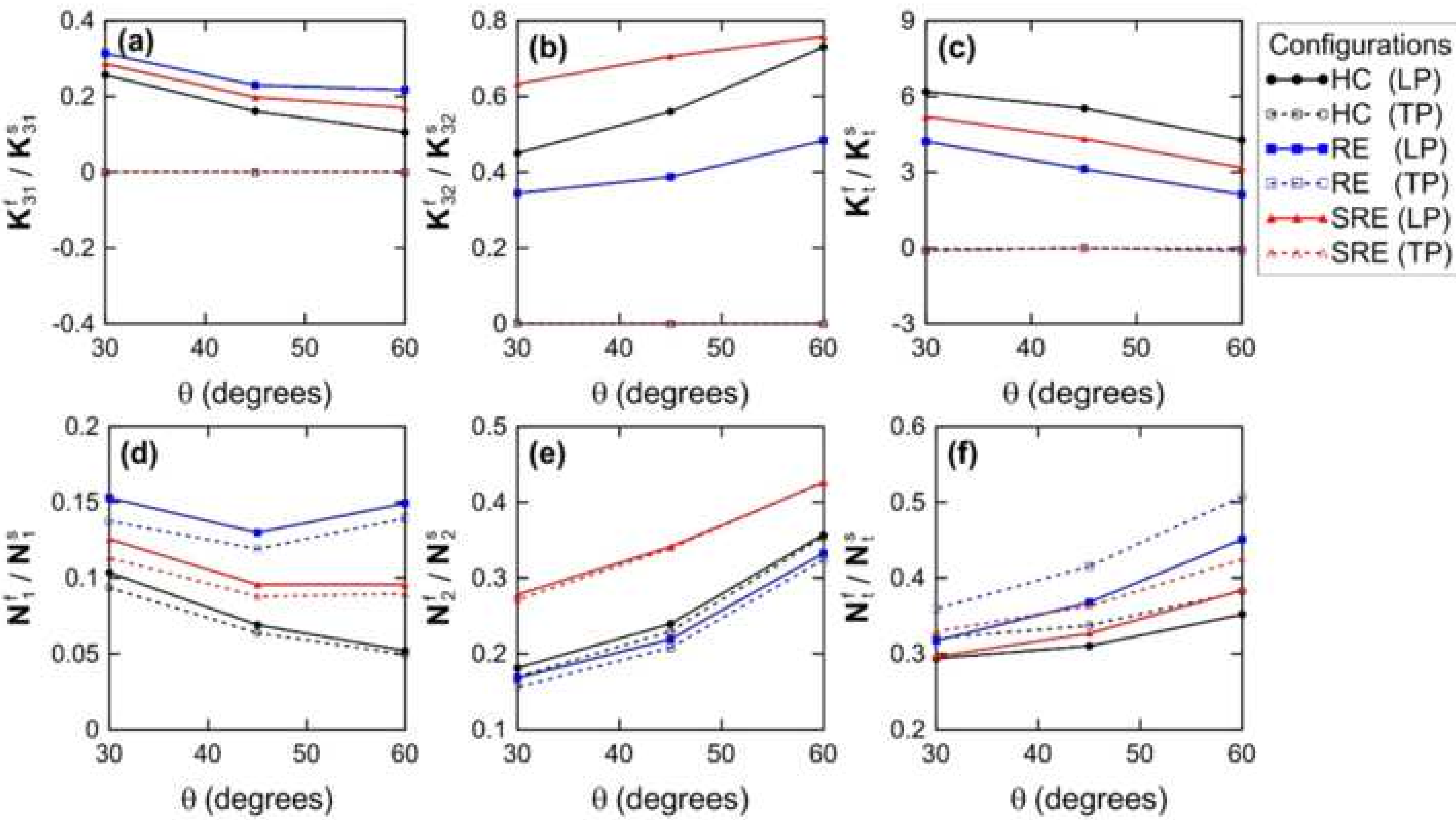

Figure 1 Piezoelectric hexagonal HC cellular networks with their unit cells a) Conventional hexagonal HC structure (CHC) b) a re-entrant HC (RE) c) semi-re-entrant HC (SRE).

Figure 2 The unit cell of HC RE meshed with 10 node quadratic tetrahedron piezoelectric elements (C3D10E) showing 6 boundary faces with respect to the axes directions.

Figure 3: Sanity Check with a) one element and b) 100x100x100 elements.

Figure *4*: The overall elastic constants of piezoelectric HC cellular network at various angles (30^o, 45^o, 60^o).

Figure 5: Variation in the overall Poisson's ratio with various angles (30^o, 45^o, 60^o) for piezoelectric HC networks.

Figure 6 Displacement and electric potential contours in the UCs of several classes of piezoelectric HC foam structures with angle 45^o. a) Displacement contours under mechanical normal loading (i.e., 0.1% normal strain along the x-axis) b) Displacement contours under mechanical shear loading (i.e., 0.1% shear strain in the x-y plane). c) Electric potential contours under electric field of 0.1MV/m.

Figure 7: The overall piezoelectric constants of HC cellular network at various angles (30^o, 45^o, 60^o).

Figure 8: The overall dielectric constants of HC cellular network at various angles (30^o, 45^o, 60^o).

Figure 9: Normalized FOMs of HC cellular network at various angles (30^o, 45^o, 60^o). (a) hydrostatic strain coefficient, (b) hydrostatic figure of merit, (c) acoustic impedance (d) Z-d_h.g_h relation

Figure 10: Selected normalized FOMs of HC cellular network at various angles (30^o, 45^o, 60^o).

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Publication Title: Piezoelectric Metamaterial with Negative and Zero Poisson's Ratio

Manuscript Title: Piezoelectric Metamaterial with Negative and Zero Poisson's Ratio

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Taryn Dollings
Editorial Coordinator
Materials Research Bulletin

January 23, 2018

Subject: Revised Manuscript Ref.: Ms. No. EMENG-4328 and responses to the reviewers' comments:

"Piezoelectric Metamaterial with Negative and Zero Poisson's Ratio"
Authors: Kamran A Khan; Sara Al Mansoor; Sohaib Zia Khan; Muhammad Ali Khan

Dear Taryn Dollings,

Enclosed with this letter, please find a detailed response to the reviewers' comments and how these comments are addressed.

Major revisions in the revised manuscript as per reviewers' suggestions:

- The introduction has been revised and many relevant and recent papers are cited with detailed discussions on
    - Why does Auxetic geometry truly matters?
    - linearized electromechanical coupled constitutive relations.

All the major modifications are highlighted with red color text.

Please do not hesitate to contact if you need any additional information.

Sincerely,

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Response to Reviewer' comments:

We would like thank the referees for their kind and constructive comments that have enabled us to clarify some important points overlooked in the original manuscript. Taking the referees' comments into consideration, we have revised the manuscript to our best. All changes are highlighted by red color text in the revised manuscript
The responses to reviewer's comments are listed below:

Comments to the Author
-Reviewer 1
This is a nice study on the Piezoelectric Metamaterial with Negative and Zero Poisson's Ratio. They used auxetic geometries to enhance the properties of piezoelectric composites. The mechanics part is nicely done. It can be published after these major comments:

Response: The authors would like to thank the reviewer for appreciating our work.

1- Piezoelectric materials are light. Why does auxectic geometry truly matter?
Response:

To address reviewer's concern on why the auxetic geometry really matter, the following paragraph has been added in the revised manuscript.

Several researchers have shown that the auxetic geometry enhances the performance of the piezocomposite and porous piezoelectric materials. Smith [1] showed that the negative Poisson's ratio polymer matrix enhances the performance of the piezocomposite materials. Numerous researchers employed topology optimization techniques and the homogenization method to improve the performance of piezocomposite materials by designing new topologies of unit cells. Sigmund et al [2] employed the topology optimization method suggested by Bendsøe and Kikuchi [3] to design 1-3 piezocomposites with optimal performance characteristics for hydrophone applications. When design for maximum d_h and $d_h.g_h$, the optimal three-dimensional, anisotropic porous matrix microstructure found to possess negative Poisson's ratios in certain directions similar to re-entrant honeycomb network. Sigmund et al [2] showed that optimized porous microstructure design enhances the values of d_h and $d_h.g_h$ over pure piezoceramics by factors of more than 10 and 10,000, respectively. Silva, Kikuchi, and co-workers [4], [5] proposed a topology optimization techniques of finding the distribution of material and voids phases in a periodic unit cell that optimizes piezocomposite electromechanical efficiency. Several porous 2D and 3D piezoelectric-polymer and piezoelectric cellular microstructure were also proposed with negative Poisson's ratio behavior.

2- Along the same line, does the geometry and cell design help strength (the enhancement is electric properties are good not magnificent)? In fact, these designs can create certain stress concentration and reduce the strength, which makes it impractical. Please comment.

Response:

For the proposed piezoelectric honeycomb cellular networks, by varying the re-entrant angle, a wide range of tunable elastic, piezoelectric and dielectric properties can be obtained that cannot be realized by monolithic piezoelectric materials. Since electromechanical properties exhibit individual dependence on an angle so they cannot all be optimized at once, though an optimum combination of properties can be obtained. For example, using topology optimization (Sigmund et al. [2]) the cell design should be optimized such that it gives the required strength and electromechanical properties. The proposed 3-1 type piezoelectric cellular networks are limited in terms of their electromechanical properties and strength. Recently, we have shown that 3-3 type piezoelectric metamaterials can provide a good combination of strength and electromechanical properties [6]. In fact, the 3-3 type piezoelectric porous metamaterials design enhances the values of d_h and $d_h.g_h$ over pure piezoceramics by factors of more than 15 and 12,000, respectively. In another study, using topology optimization, Sigmund et al [2] also showed that optimized porous microstructure design enhances the values of d_h and $d_h.g_h$ over pure piezoceramics by factors of more than 10 and 10,000, respectively.

This is true that the cellular lattice materials are prone to stress concentration regions but as discussed earlier the compromise in strength and electromechanical properties should be sought by the designer for the specific application and select the optimized design of cellular structure accordingly.

-Reviewer 2

Khan presents a finite-element based micromechanical modeling framework to compute the electromechanical properties of a series of periodic piezoelectric materials. Negative and zero Poisson's ratios were predicted based on averaged model. The work done in the paper is extensive, and the paper is well written with some revisions to be made before publication:

1. There are numerous grammatical errors and mistakes made in the paper.
   For example, line 101, it should be "described",
   line 11, "to" is to be removed,
   line 155, "total (of) four degree(s) of freedom..." is the correct form, and 2 commas are needed, one before "three" and one before "and",
   line 175 must be "poled" not "pole".
   Other lines that need corrections, among others, are line 196, 224, 257 (the figure number), 307, 308, 386, 387, 389, etc.

Response:

We appreciate the reviewer's careful review and thankful for suggested corrections. All corrections have been made. Manuscript is carefully revised with the help of an English language expert.

2. Authors have used linearized electromechanical coupled constitutive relations for the piezoelectric material being used. This formulation is generally accepted. Thereafter, the authors assume a similar form of constitutive relation, with averaged values, for the RVE that is build from the piezoelectric unit cell. This assumption leads to an orthotropic linear material model. We already know from the literature [1,2, 3] that linear microelements in an RVE will produce nonlinear behavior of the macro-material point. Even in the simplest case for the geometry shown in the paper, and for a linear isotropic material (and not a piezoelectric material), the geometry-driven non-linearity prevents the use of linear models as linear models do not correctly exhibit the complexity and inter-connectivity of different parameters involved in formulating the problem. Perhaps, a justification of using Eq. 2 in the paper will answer such objectives.

[1] Misra, A. and P. Poorsolhjouy, Granular micromechanics based micromorphic model predicts frequency band gaps. Continuum Mechanics and Thermodynamics, 2016. 28(1-2): p. 215-234.
[2] Rosi, G. and N. Auffray, Anisotropic and dispersive wave propagation within strain-gradient framework. Wave Motion, 2016. 63: p. 120-134.
[3] dell'Isola, F., et al., Pantographic metamaterials: an example of mathematically driven design and of its technological challenges. Continuum Mechanics and Thermodynamics, 2018: p. 1-34.

Response:

Thanks for providing the above references. We appreciate the reviewer's recommendation and found it useful for our research. To address the reviewer's concern, we have added a small paragraph discussing the role of linearized electromechanical coupled constitutive relations in designing piezocomposites and porous piezoelectric materials. We have cited the above mentioned reference and many relevant and recent papers in the revised manuscript.

To address reviewer's concern, the following text has been added in the revised manuscript.

We considered the linearized electromechanical coupled constitutive relations for each ligament material to obtain the homogenization electromechanical properties of the proposed cellular network. These linearized constitutive relations are simple, attractive and, arguably, the most feasible approach for describing the electromechanical response of a cellular network at the macroscale [7],[8],[9],[2]. These assumptions lead to an orthotropic linear electromechanical material model. However, there are advanced models available in the literature [[10], [11], [12]] that showed linear microelements in an RVE will produce a nonlinear homogenized response. The geometry-driven non-linearity prevents the use of linear models as linear models do not correctly exhibit the complexity and inter-connectivity of different parameters involved in formulating the problem. In the present study, we do not consider models that account for geometry-driven nonlinearity. However, it will be interesting to analyze the proposed cellular network using such advanced models that account for higher order gradient theory and geometry-driven non-linearity.

3. A consequence of the point mentioned in (2) is that the behavior of the material when excited will not be dispersive, while we know from [1] that dispersion occurs and the velocity of wave will be a function of the frequency/wavenumber. An explanation on this matter will also help the reader in grasping the physics of the problem.

Response:

The author's would like to thank the reviewer for raising this interesting point. The following text has been added in the revised manuscript discussing the behavior of material subjected to excitation while considering higher order gradient continuum models.

It should be emphasized here that the use of linearized electromechanical coupled constitutive relations has some implications. For example, higher order gradient continuum models have been recently used by several researchers to investigate the dispersive wave propagation in strain gradient elastic media. Contrary to classical elasticity within the strain gradient framework, Rosi and Auffray [11] showed that the wave propagation in hexagonal lattices becomes anisotropic and group velocity was proven to be different from energy velocity and should be treated as different quantities. Misra and Poorsolhjouy [10] derived a micro-morphic continuum model for the elasticity of granular media and the dispersion graphs have presented showing the relationship between the micro-scale parameters and the dispersion behavior. Contrary to classical elasticity where all waves will be of an acoustic type and there will be no possibility of frequency band gaps, the micro-morphic continuum model has the capability to present band gaps over a large range of wave numbers. Their results indicate that there is a possibility of designing materials with specific wave propagation behaviors that can be used as alternates to piezoelectric materials used commonly for structural vibration control or for damage identification.

.

4. Line 185-186, how is forcing parallel faces of the unit cell to remain parallel justified? This extra boundary condition must be accounted for in the energy expressions. I suggest the authors clarify such an assumption further by stating the pros and cons of such an assumption.

Response:

We feel that this sentence is confusing and it does not explains clearly what we used in our computation. Here, the forcing of parallel faces of the unit cell to remain parallel is referred to only the faces subjected to displacement loading along one specific degree of freedom for load cases 1-6. The other faces follows the deformation mechanism as per the boundary conditions [13],[14],[15].

On the loading face, we use the common concept of master node to apply the boundary conditions. Here, the master node (the loading direction, e.g., displacement along x-axis for load case 1) is coupled with all the nodes on respective loading face along one degree of freedom. This coupling allows all the nodes to be displaced by the same amount as a master node. For the loading cases 7-9 there is no such conditions of forcing of parallel faces of the unit cell to remain parallel is considered.

To avoid confusion, we have removed the sentence on line 185-186.

Enforcing any additional boundary condition that has not been accounted in energy expression may lead to over- or under-stiff homogenized behavior of the unit cell and incorrect micro field variables distribution.

[1] W.A. Smith, Optimizing electromechanical coupling in piezocomposites using polymers with negative Poisson's ratio, in: IEEE 1991 Ultrason. Symp., 1991: pp. 661–666 vol.1. doi:10.1109/ULTSYM.1991.234109.
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