

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

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

32 **Introduction**

33
34 Piezoelectric materials (PMs) play a key role in advanced multifunctional composites industry by
35 virtue of their unique electromechanical coupling characteristics (Muliana 2011). PMs have found
36 applications in actuators, sensors, ultrasound imagers, hydrophones and echo-cardiogram
37 (Marselli et al. 1999). In these devices, PMs convert mechanical energy into electrical or vice-
38 versa. For example, in sensing applications, PMs require high sensitivity and low acoustic
39 impedance (Alkhader et al. 2015; Lethiecq et al. 2004). Existing bulk piezoelectric polymers show
40 low acoustic impedance and sensitivity while ceramics type piezoelectric materials have high
41 acoustic impedance and sensitivity (Kar-Gupta and Venkatesh 2006). Several studies were
42 conducted to enrich the coupling characteristics between the mechanical and electrical properties
43 in piezoelectric materials by either embedding constituents (Topolov and Bowen 2008) or by
44 introducing the porosity (Hikita et al. 1983).

45 Piezoelectric composites were tailored to exhibit improved piezoelectric activity and mechanical
46 flexibility (for example, active fiber composites, AFM) (Elhajjar et al. 2013). However, the
47 porosity can significantly enhance the performances of medical diagnostic devices (Smith 1989)
48 and hydrophones (Geis et al. 2000). Both piezoelectric composites (e.g., (Skinner et al. 1978);
49 (Ramesh et al. 2006); (Richard et al. 2004)), and porous PMs (e.g., Nagata et al. 1980; Hikita et
50 al. 1983; Ting 1985; Bast and Wersing 1989; Bowen et al. 2004; Piazza et al. 2005; Zhang et al.
51 2007; Lee et al. 2007) showed potential in obtaining low acoustic impedance and high piezoelectric

52 sensitivity. However, currently, piezoelectric composites demonstrate properties far from the ideal
53 electromechanical characteristics.

54 In designing the porous PMs and the piezoelectric composites, the spatial distribution of the two
55 phases controls the effective enhancement of the electromechanical properties (Levassort et al.
56 1998). Therefore, the concept of phase connectivity was defined (Newnham et al. 1978). To
57 understand the role of connectivity of porosity on the properties of PMs, several experimental
58 studies were conducted by considering numerous configurations of porosity, such as, embedded
59 porosity in a PMs (3-0 foam) (Marselli et al. 1999; Kara et al. 2003; Ueda et al. 2010); long
60 cylinder-like porosity (3-1 foam) (Wirges et al. 2007) and open-cell like porosity (3-3 foam)
61 (Nagata et al. 1980). Results demonstrated that porosity help to enhance the sensitivity of PMs.

62 Several analytical studies were conducted to estimate the electromechanical characteristics of
63 porous PMs by considering simplified configurations of porosity (such as 3-0 and 3-1) (Banno
64 1985, Dunn and Taya 1993b, Mikata 2001, Bowen and Topolov 2003). Several micromechanical
65 models based finite element frameworks were developed for predicting electromechanical
66 properties of 3-0, 3-1 and 3-3 type porous PMs and hence addressed more complex microstructure
67 (Kar-Gupta and Venkatesh 2008, Bosse et al. 2012). These studies established that behavior of
68 porous PMs governed by the porosity level, pore shape, pore direction, and their distribution,
69 cellular interconnectivity and the direction of the poling (Iyer et al. 2015).

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

74

75

76 Among Piezoelectric cellular solids and architected foams, the 3-1 type cellular honeycomb
77 (HC) configuration is highly in use due to its simplicity, utility, and workability with structures.
78 Based on the ligament orientations the HC architecture may produce deformation with positive
79 (conventional), negative (auxetic) and zero Poisson ratio's (Grima et al. 2010). Auxetic materials
80 are attractive due to their counterintuitive response under deformation and improved properties.
81 The structural behavior of HC cellular networks with passive elastic anisotropic ligaments
82 representing auxetic effects has been extensively studied (Grima et al. 2010, Masters and Evans
83 1996, Gibson and Ashby 1997, Zhu et al. 1997). These studies established that the ligament
84 orientation of cellular network governed the hexagonal HC architecture-property relationship
85 (Alkhader and Vural 2009; Papka and Kyriakides 1999).

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

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

100 **Architecture of Piezoelectric Cellular Material**

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

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

115 **Constitutive Model of Piezoelectric Cellular Material**

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

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

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

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

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

130 We considered the linearized electromechanical coupled constitutive relations for each ligament
 131 material to obtain the homogenized electromechanical properties of the proposed cellular network.
 132 These linearized constitutive relations are simple and, arguably, the most practical method for
 133 describing the electromechanical behavior of cellular network at the macroscale (Challagulla and
 134 Venkatesh 2013, Iyer et al. 2014, Kar-Gupta and Venkatesh 2006, Sigmund et al. 1998). These
 135 assumptions lead to an orthotropic linear electromechanical material model. However, there are
 136 advanced models available in the literature (Misra and Poorsolhjouy 2016, Rosi and Auffray 2016,
 137 Dell'Isola et al. 2018) that showed that linear microelements in an RVE would produce a nonlinear
 138 homogenized response. The geometry-driven non-linearity prevents the use of linear models as

139 linear models do not correctly exhibit the complexity and inter-connectivity of different parameters
140 involved in formulating the problem. In the present study, we do not consider models that account
141 for geometry-driven nonlinearity. However, it will be interesting to analyze the proposed cellular
142 network using such advanced models that account for higher order gradient theory and geometry-
143 driven non-linearity.

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

158 **Micromechanical Finite Element Model for Piezoelectric Cellular Materials**

159 Recently, Khan and Abu Al-Rub (Khan and Abu Al-Rub 2017, 2018a; b) developed an FE-
160 based UC homogenization method to predict the overall mechanical properties of periodic
161 architected materials based on the microstructure geometry and its base material properties. In

162 this study, we have extended the framework to calculate the overall properties of architected and
163 periodic cellular PMs. Figure 1 shows the UCs of the three types of hexagonal HC structures.
164 Periodic boundary conditions (PBCs) are imposed on UC (Luxner et al. 2005, Kanit et al. 2003;
165 Khan and Muliana 2009, Choudhry et al. 2016) that yield the responses of infinitely repeating
166 patterns of architecture (Jiang et al. 2002, Abueidda et al. 2015). PBCs give quite reasonable
167 estimates of the properties as compared to the homogeneous displacement and homogeneous
168 traction boundary conditions (Zohdi and Wriggers 2005, Xia et al. 2003). Using the proposed
169 framework, it is possible to characterize the linear electromechanical response of piezoelectric
170 architecture materials completely.

171 **Finite Element Models**

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

182 The cellular materials were assumed to be poled longitudinally and transversely. We have
183 assumed that every region of the FE model considered in this study was poled uniformly in one
184 preferred direction. Uniform poling of the piezoelectric HCs normal to transverse porosity

185 direction is a very challenging task. However, fabrication of the cellular HCs porous networks
 186 with uniform poling along the longitudinal direction has been successfully demonstrated (Fey et
 187 al. 2016).

188
 189 **Boundary conditions and homogenization for RVE**

190 PBCs are usually applied to the UC to ensure that UC represents the response of the whole
 191 architected foam. Moreover, the displacements compatibility and the electric potential continuity
 192 across neighboring UC boundaries are assured (Iyer and Venkatesh 2010, Iyer and Venkatesh
 193 2011).

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

$$196 \quad u_i^{j+}(x, y, z) - u_i^{j-}(x, y, z) = c_i^j \quad (i, j = 1, 2, 3) \quad (3)$$

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

$$199 \quad \phi^{j+}(x, y, z) - \phi^{j-}(x, y, z) = \bar{E}_i (x_i^{j+} - x_i^{j-}) \quad (i = 1, 2, 3) \quad (4)$$

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

202 One of the requirements of the Eq. (3) is that on the two opposite boundary surfaces, the difference
 203 of the displacements of the corresponding points should be specified. However, Li (Li 2008)
 204 derived an explicit displacement BCs of UCs representing microstructure of periodic structure
 205 taken from the symmetry existent within the structure. In this study, the Li (Li 2008) work has
 206 been extended to account for electric charge continuity across neighboring UCs and the mixed

207 boundary conditions are proposed to compute the overall properties of piezoelectric architected
 208 materials. Table 2 shows the list of the B.C.s.

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

$$212 \quad \bar{\sigma}_{ij} = \frac{1}{V} \int_V \sigma_{ij}(x, y, z) dV \quad \bar{\varepsilon}_{ij} = \frac{1}{V} \int_V \varepsilon_{ij}(x, y, z) dV \quad (5)$$

213 Analogously the average electric fields and electric displacements are defined by

$$214 \quad \bar{E}_i = \frac{1}{V} \int_V E_i(x, y, z) dV \quad \bar{D}_i = \frac{1}{V} \int_V D_i(x, y, z) dV \quad (6)$$

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

$$216 \quad \bar{\sigma}_{ij} = \frac{R_{ij}}{A_j} \quad (\text{no summation on } j) \quad (7)$$

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

$$218 \quad \bar{D}_i = \frac{q_i}{A_i} \quad (8)$$

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

223 **Sanity Check**

224 In the literature, there is yet no analytical formulation available that offer explicit computation of
 225 the properties of the piezoelectric HC networks while their ligaments following orthotropic

226 material behavior. Hence, the mixed boundary conditions given in Table 2 was applied to one
 227 element and 100x100x100 elements cube to evaluate the correctness and appropriateness of the
 228 proposed methods. It was found that the proposed approach yields the homogenous properties of
 229 the PZT-5H given in Table 1.

230 **Results and Discussion**

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

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

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

245 Where $[C^D]$ and $[S^D]$ are the components of the stiffness and compliance tensor measured at
 246 zero electric displacements, respectively. The $[e]$, $[h]$, $[g]$ are the piezoelectric stress tensor,

247 strain tensor, and voltage tensor, respectively. The dielectric strain tensor $[\kappa^{\mathcal{E}}]$ is computed at
 248 zero strain.

249
 250 **Effective Elastic Response**

251 The Compliance matrix coefficients \bar{S}_{ijkl}^E were used to calculate the stiffness constants \bar{C}_{ijkl}^E .

252 **Figure 3** illustrates the stiffness constants of longitudinally and TP HC structures over various
 253 angles 30° , 45° , and 60° . Both LP and TP HC cellular materials showed substantial variations in
 254 both in-plane and out-of-plane stiffness coefficients values for different θ . The elastic properties
 255 were found to be anisotropic and showed different trends and non-intuitive behavior.

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

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

267 However, the out-of-plane normal stiffness constant C_{13} showed higher stiffness as compared
 268 to the in-plane stiffness constant C_{12} . The in-plane compliant behavior is due to the fact that the
 269 in-plane Poisson's ratio of the HC network is about one order of magnitude higher than the out of

270 plane Poisson's ratio. For example, the in- (ν_{12}) and out-of- (ν_{13}) plane Poisson's ratio of the
271 conventional HC with an angle of ligaments of 30° is 0.44 and 0.034, respectively.

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

288 **Figure 6** shows the displacement and electric potential contour plots for three different load case.
289 For all three HC structures considered, the displacement contours in 1-direction under uniaxial and
290 shear loading loadings are shown in **Figures 6(a)** and **(b)**, respectively. These distributions of
291 displacement fields clearly demonstrated the state of shear and uniaxial deformation behavior. For
292 the loading case-7, the electric field of 0.1MV/m is applied, and the electric potential contours

293 plots are obtained. **Figure 6(c)** shows that for all three HC network, there is a linear variation of
294 electrical potential between opposite faces of loading while a uniform electric potential was
295 observed at the central part of the UCs.

296 **Effective piezoelectric properties**

297

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

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

312 The effective normal piezoelectric properties were found to be a function of ligament angle and
313 HC networks architecture. The normal piezoelectric constant, e_{33} , is usually recognized as the most
314 vital parameter for various applications utilizing monolithic PMs. For LP HC network e_{33} (LP) was
315 amplified with the increase in ligament angle, but there was an insignificant variation for e_{33} (TP).
316 In addition, **Figure 7** demonstrated that even though the e_{31} of the ligament base material has

317 negative values, the HC network could display both -ve and +ve values of e_{31} values by varying
 318 the angle θ . These results indicated that the piezoelectric HC architecture could show a different
 319 crystal symmetry as compared to the ligament base material. The LP HC network produced the
 320 highest negative values of e_{31} and e_{32} .

321 **Effective dielectric properties**

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

330 **Effective Figure of Merit**

331 Various figures of merit (FOM) are of interest to evaluate the usefulness of PMs in industrial
 332 applications, such as an ultrasound imager, hydrophones, and energy harvesters. The combination
 333 of fundamental electromechanical coefficients (i.e., elastic, piezoelectric and dielectric coefficients
 334 as calculated by the above mentioned FEA) can be used to evaluate numerous industrial FOM. For
 335 cellular network FOM, the relevant constants in various applications (i.e., hydrophones) are the
 336 hydrostatic strain coefficient (d_h), the hydrostatic FOM ($d_h \cdot g_h$), the acoustic impedance (Z) and
 337 electromechanical thickness model coupling factor (k_t) (Kar-Gupta and Venkatesh 2006, Dunn
 338 and Taya 1993b). There are some other FOM which are of importance in several different

339 applications. The relevant FOM parameters are given in Table 3. More details on FOMs can be
 340 found elsewhere, (Dunn and Taya 1993b).

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

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

357 The Z - $d_h \cdot g_h$ relationship is important to be analyzed while designing the transducer and
 358 hydrophones. **Figure 9** shows the Z - $d_h \cdot g_h$ quid pro quo, where when the porosity of HC networks
 359 increases, the $d_h \cdot g_h$ showed increasing trend while the Z value decreases. Such a quite unique
 360 combination of increased sensitivity and reduced acoustic impedances is nearly impossible to

361 obtain using bulk PMs and piezoelectric composites (Dunn and Taya 1993a). These results indicate
362 that it is possible to design architected porous PM with required electromechanical characteristics.
363 Several researchers have shown that auxetic geometry improves the performance of the
364 piezoelectric composite and porous PMs. For piezoelectric composites, Smith (Smith 1991)
365 showed that the polymer matrix with negative Poisson's ratio enhances their performance.
366 Numerous studies combined topology optimization and finite element methods to design optimum
367 topologies of unit cells to obtain a piezoelectric composite with better behavior. Using the topology
368 optimization method proposed by Bendsøe and Kikuchi (Bendsøe and Kikuchi 1988), the optimum
369 design of 1-3 piezoelectric composites for hydrophone applications were obtained by Sigmund et
370 al (Sigmund et al. 1998). Using the criteria to maximize for d_h and $d_h \cdot g_h$, the obtained optimal
371 3D porous architected matrix showed auxetic effects in certain directions like re-entrant type HC
372 cellular material. Moreover, the optimized design with porous architecture design increases the
373 values of d_h and $d_h \cdot g_h$ over monolithic piezoelectric ceramics by factors of more than 10 and
374 10,000, respectively. A unit cell based topology optimization approach was proposed by Silva,
375 Kikuchi, and co-workers (Silva et al. 1997, Silva et al. 1998) to find the distribution of inclusion
376 and/or voids phases that enhances piezoelectric electromechanical efficiency. Several 2D and 3D
377 auxetic structures (negative Poisson's ratio) consisting of piezoelectric polymer and architected
378 cellular materials were proposed.

379 For the proposed piezoelectric HC cellular networks, a desirable elastic, piezoelectric and
380 dielectric properties can be obtained that is nearly impossible to achieve using bulk PMs and
381 piezoelectric composites. The unique set of electromechanical properties is a function of angle so
382 all the properties cannot be improved at once. It requires some techniques to obtain an optimum
383 combination of properties. For example, using topology optimization Sigmund et al. (Sigmund et

384 al. 1998) showed that the cell design should be optimized such that it gives the required strength
 385 and electromechanical properties. The proposed 3-1 type piezoelectric cellular networks are
 386 limited in terms of their electromechanical properties and strength. Recently, we have shown that
 387 3-3 type piezoelectric metamaterials can provide a good combination of strength and
 388 electromechanical properties (Khan and Khan 2019). The 3-3 type piezoelectric porous
 389 metamaterials design improves the values of d_h and $d_h \cdot g_h$ over monolithic piezoelectric ceramics
 390 by factors of more than 15 and 12,000, respectively.

391 Next, we analyzed the behavior of normalized electromechanical coupling factors (k_{31}, k_{32}, k_t)
 392 and frequency constants (N_{31}, N_{32}, N_t). **Figures** 10(a-c) showed the normalized magnitudes of
 393 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}
 394 were equal for the bulk PZT-5H. For all proposed HC cellular materials, for given angles, the k_{31}
 395 values were seen to decrease more comparative to k_{32} because of the anisotropic nature of
 396 microstructure. For both modes, the d_{3i} were nearly equal but the higher values of S_{11}^E than S_{22}^E
 397 give rise to more decrease in k_{31} than k_{32} . In contrast, to k_{31} and k_{32} , the normalized k_t values were
 398 one order higher than the monolithic PZT-5H.

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

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

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

419 **Conclusions**

420 This study proposed a finite element base micromechanical modeling framework to estimate
421 the elastic, dielectric and piezoelectric of the HC based cellular PMs. Using the internal symmetry
422 of the periodic structure a mixed boundary conditions analogous to PBCs were proposed. The
423 modeling approach was applied to the UCs of conventional, auxetic and semi re-entrant type HC
424 piezoelectric cellular networks and their electromechanical properties were presented. For the
425 longitudinal poled network, the results showed that the HC network exhibited an exceptional
426 combination of piezoelectric properties, i.e., low impedance and more sensitivity, which could not
427 be obtained by monolithic PMs. However, for the TP network, the electromechanical properties
428 displayed insignificant dependence on the porosity and angle of the ligament. The FE results

429 showed that HC network with tunable electromechanical characteristics coupled with auxetic
430 behavior such as negative or zero Poisson's ratio could be produced. Such novel HC network-
431 based functional cellular materials have the capacity to facilitate the design of light-weight devices
432 for various next-generation sensors and actuators.

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625

1 **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 | 4 |
| $S_{12}^E = S_{21}^E$ (pm ² / N) | -4.78 | |
| $S_{13}^E = S_{23}^E$ (pm ² / N) | -8.45 | 5 |
| S_{33}^E (pm ² / N) | 20.7 | 6 |
| S_{44}^E (pm ² / N) | 42.6 | 7 |
| $S_{55}^E = S_{66}^E$ (pm ² / N) | 43.5 | |
| $d_{15} = d_{24}$ (p m / V) | 741 | 8 |
| $d_{31} = d_{32}$ (p m / V) | -274 | |
| d_{33} (p m / V) | 593 | 9 |
| $\kappa_{11}^\sigma / \kappa_0 = \kappa_{22}^\sigma / \kappa_0$ | 3130 | 10 |
| $\kappa_{33}^\sigma / \kappa_0$ | 3400 | |
| Density (ρ) | 7500 kg/m ³ | 11 |
| Permittivity of free space (κ_0) | 8.85x10 ⁻¹² C/Vm | 12 |

13

14

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

17

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

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

20

21

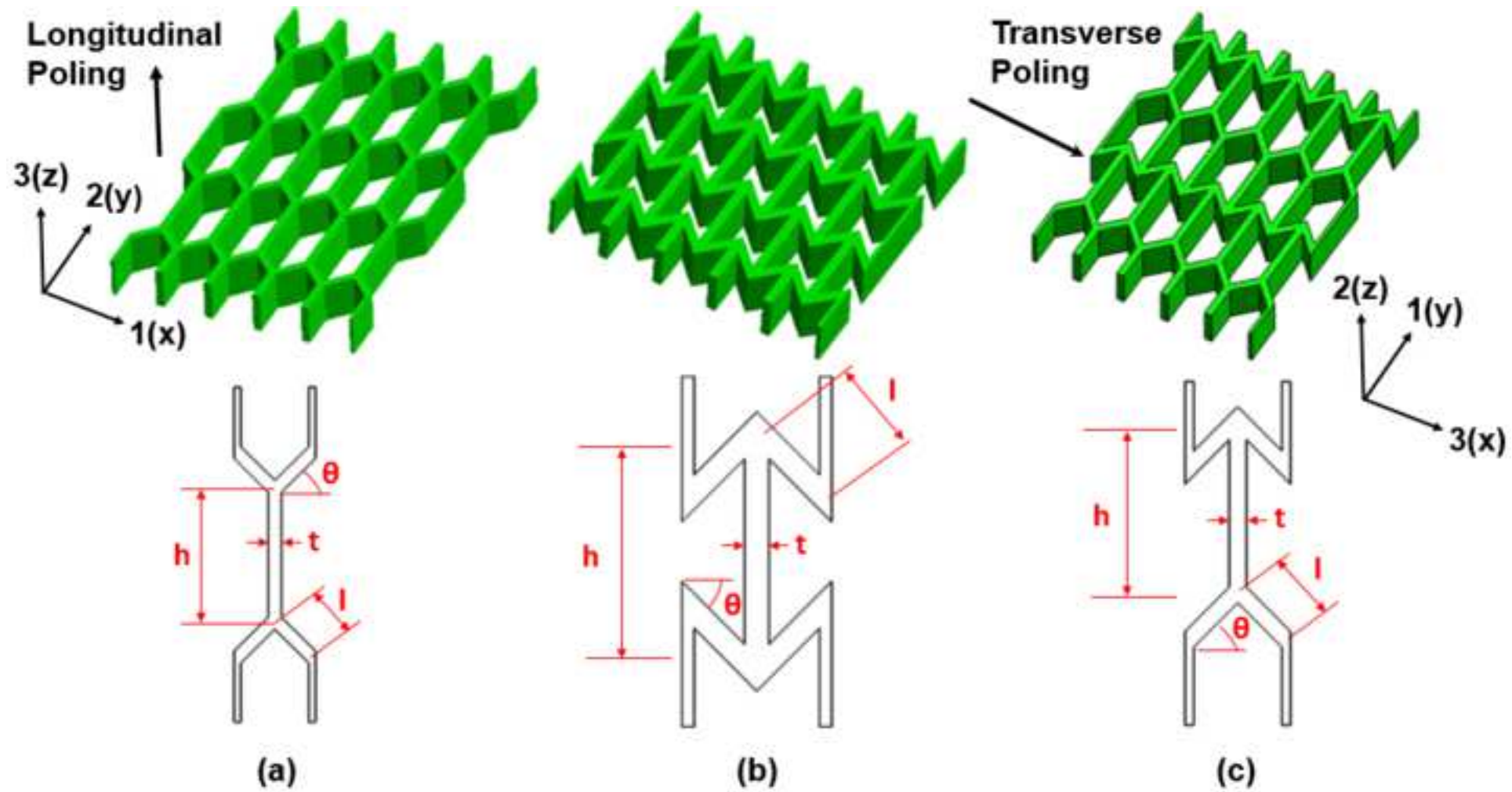
22 **Table 3. List of figure of merit.**
 23

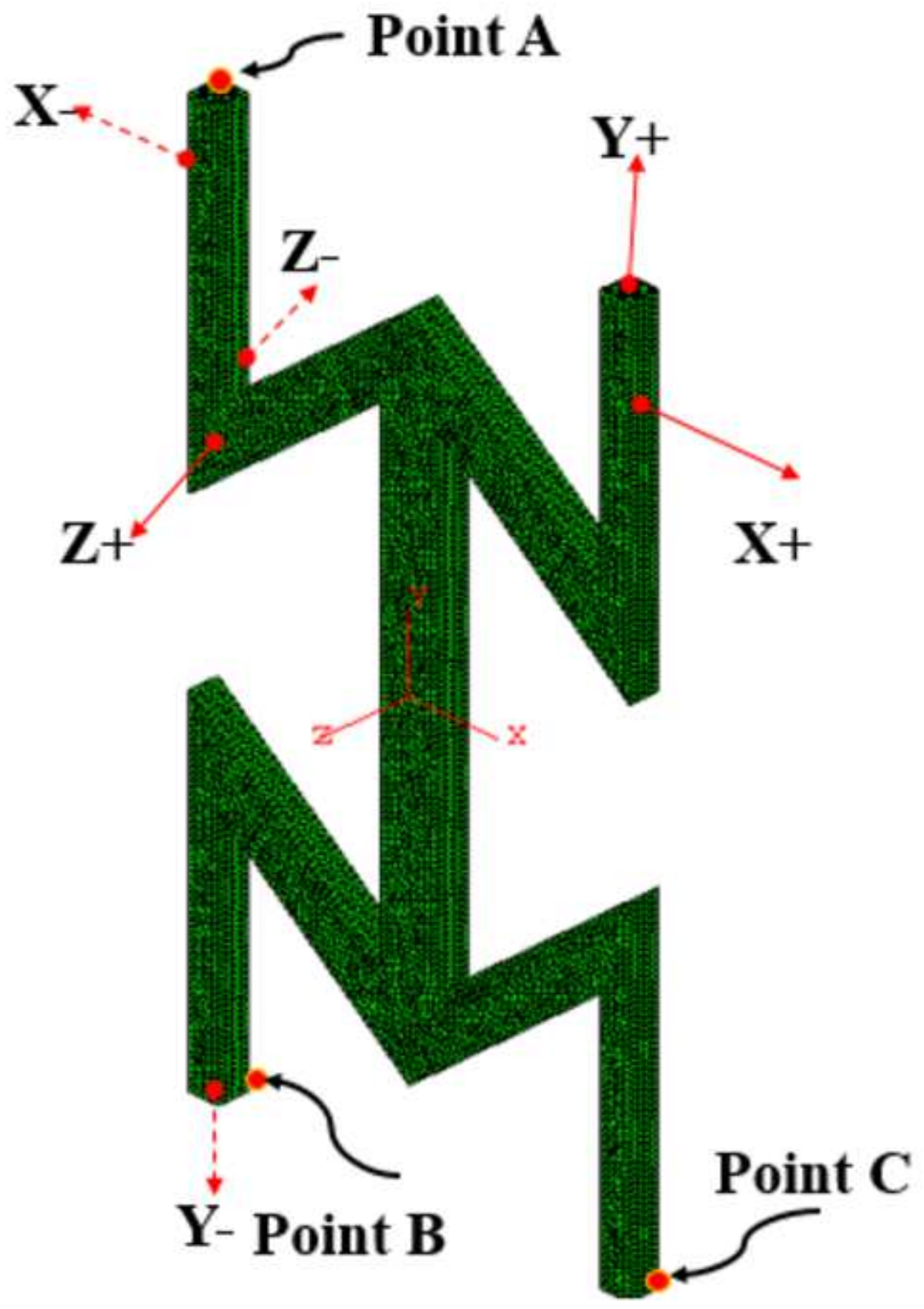
| Parameter | Relation |
|------------------------------------------------------------|------------------------------------------------------------------------------------------------------------|
| hydrostatic strain coefficient (d_h) | $d_h = d_{31} + d_{32} + d_{33}$ |
| hydrostatic figure of merit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

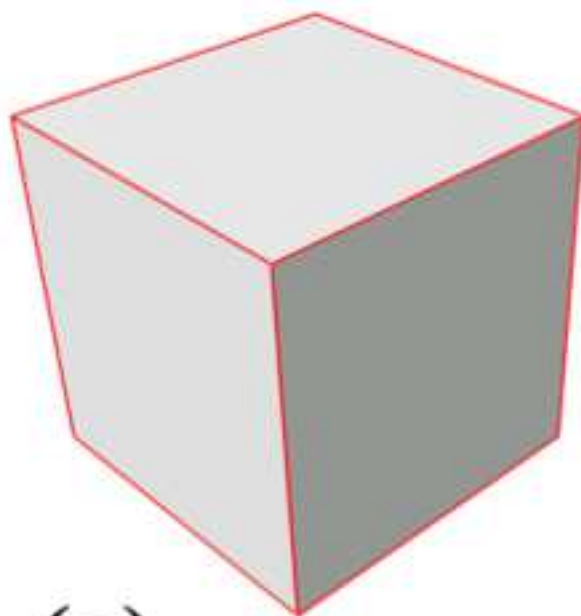
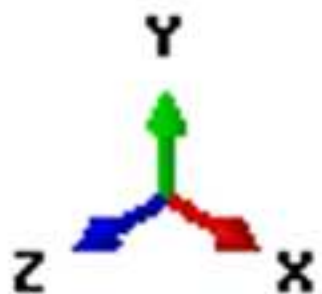
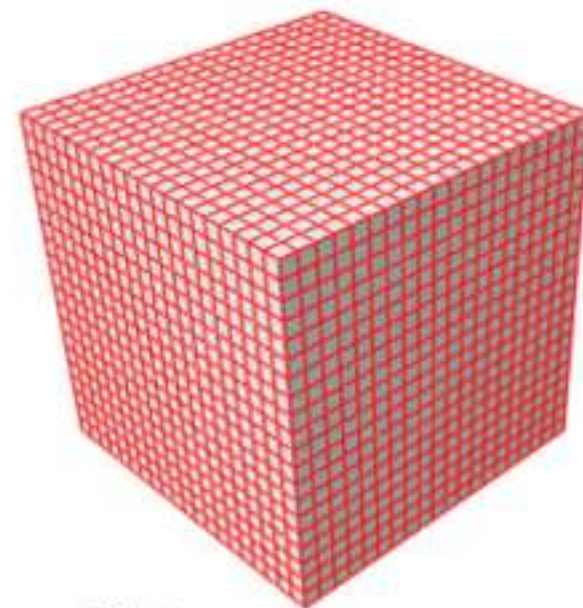
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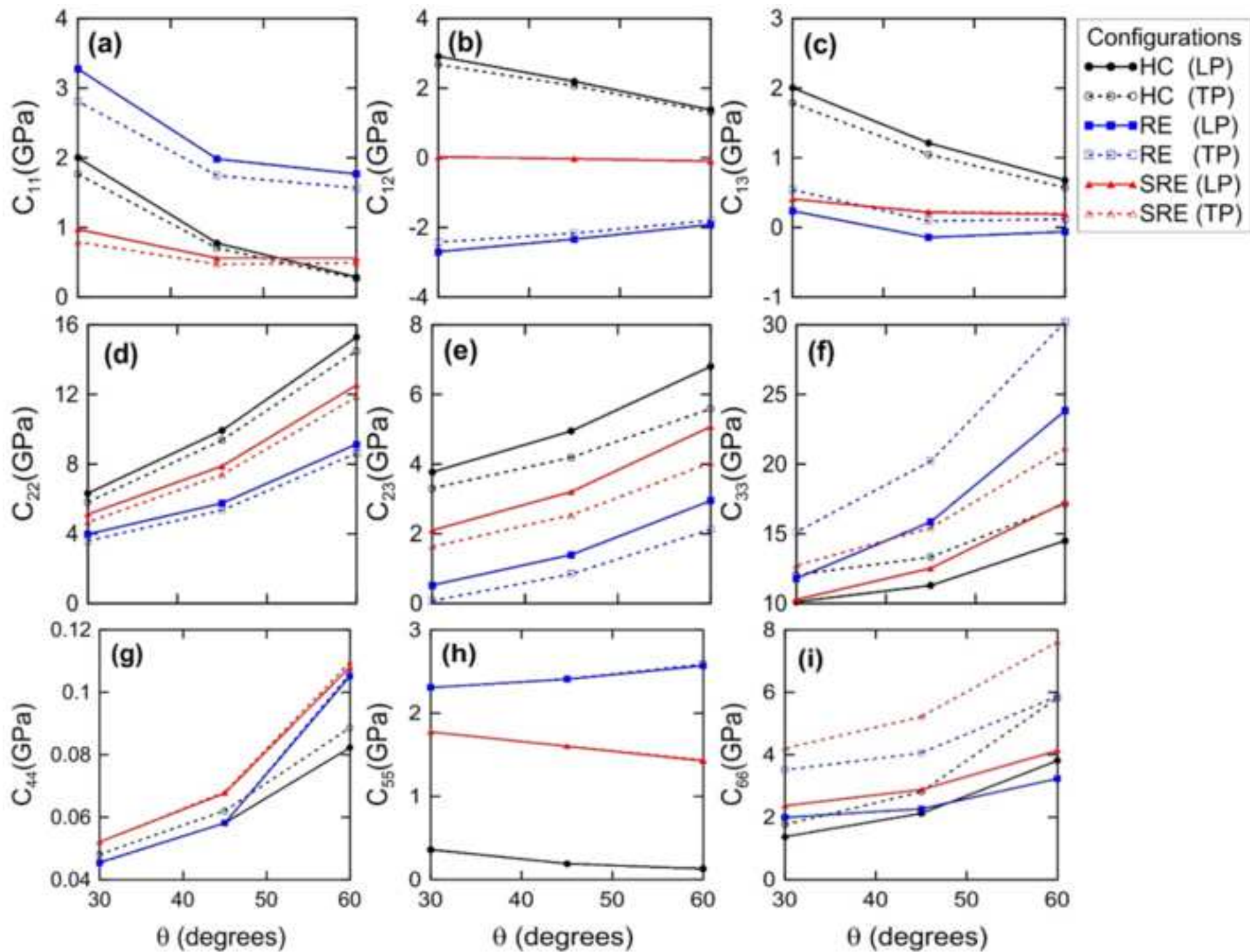
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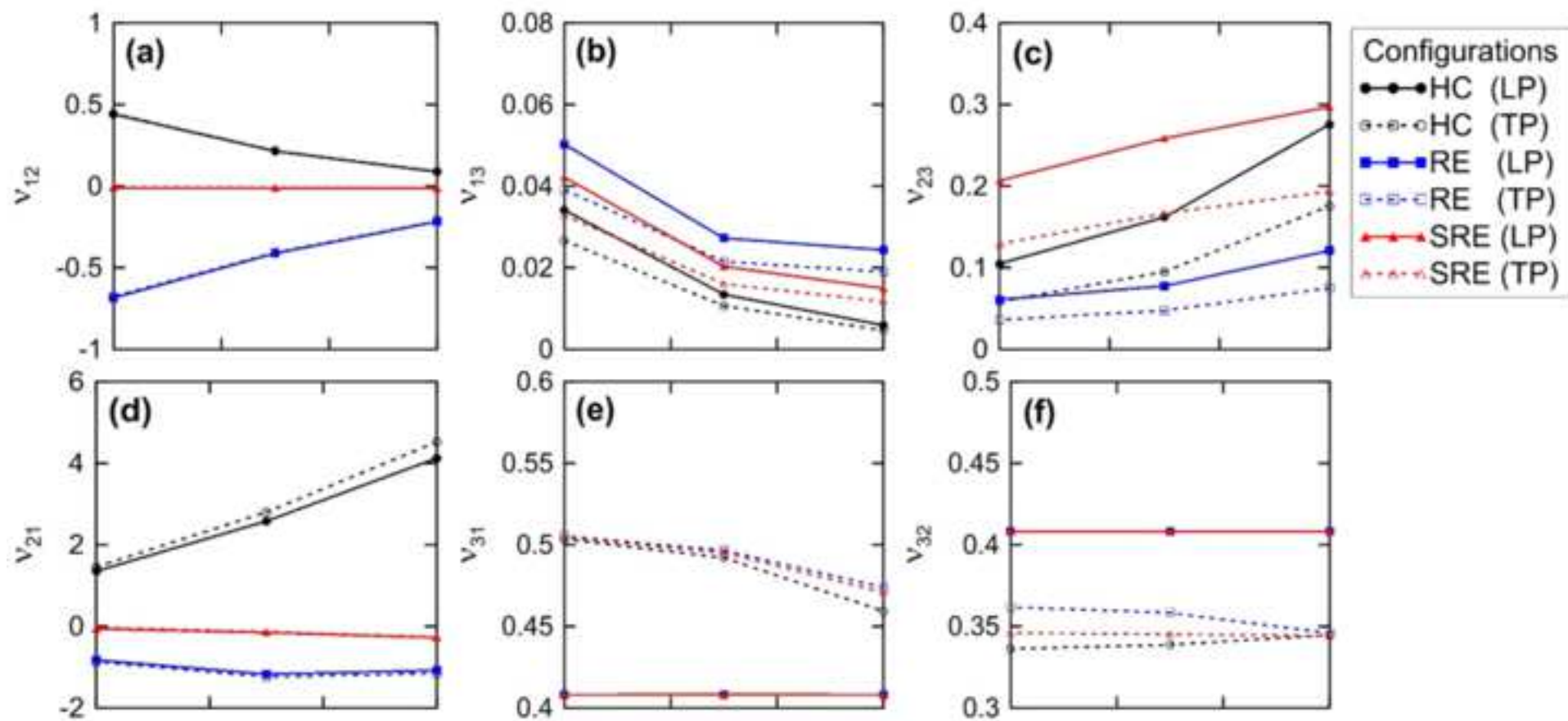
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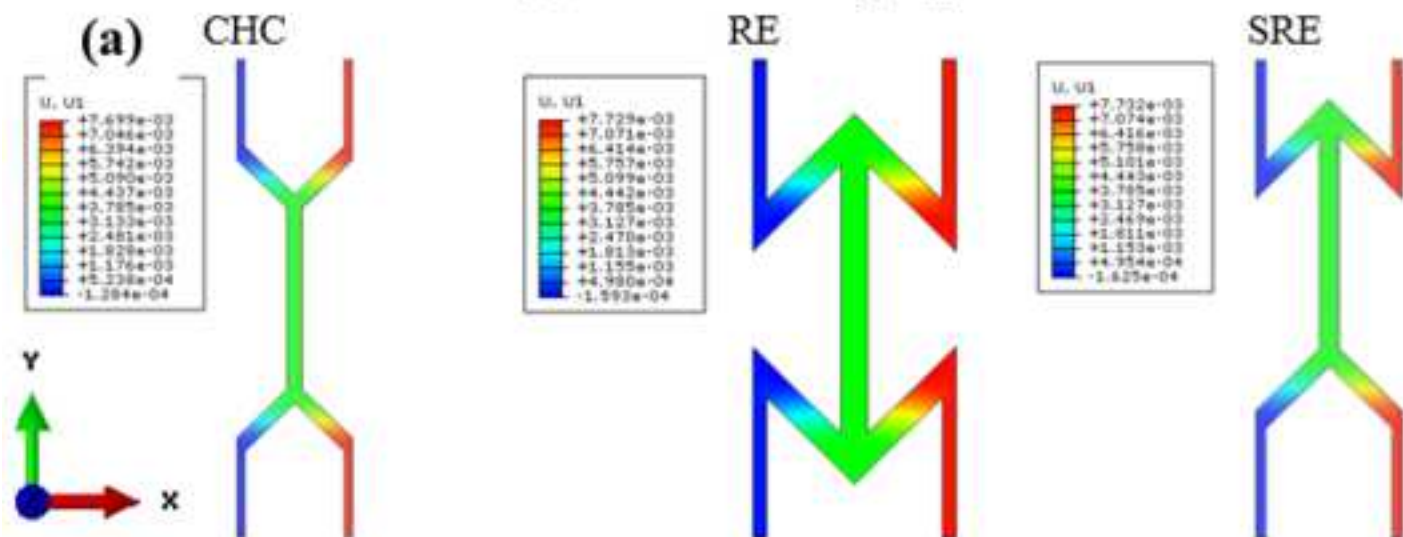
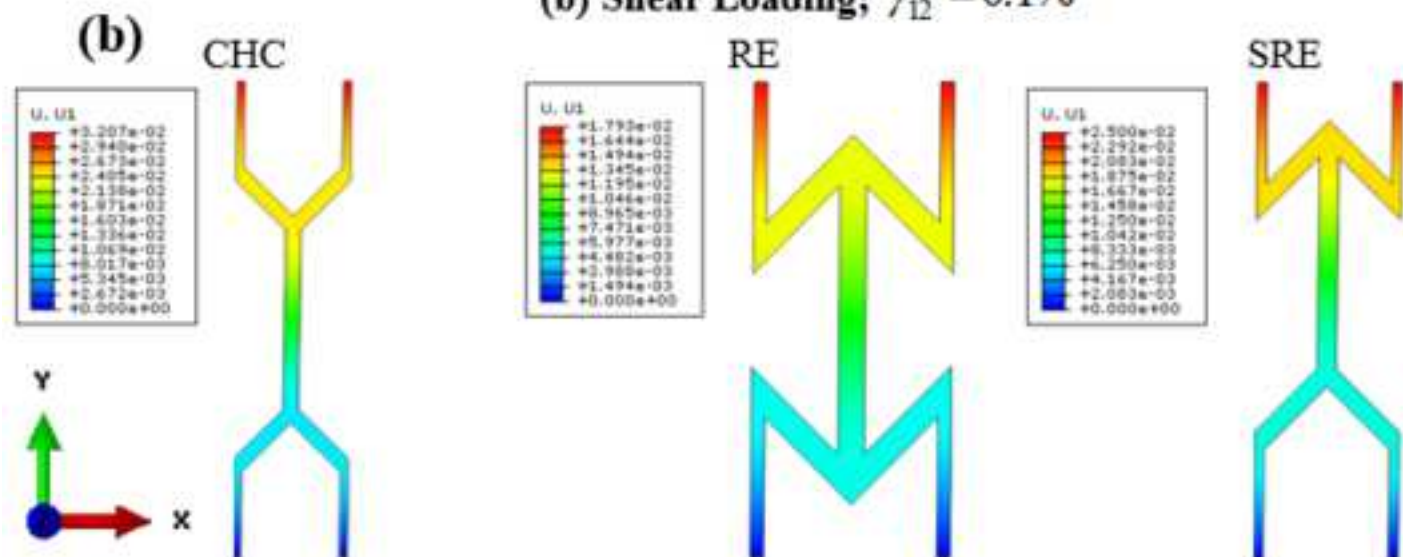
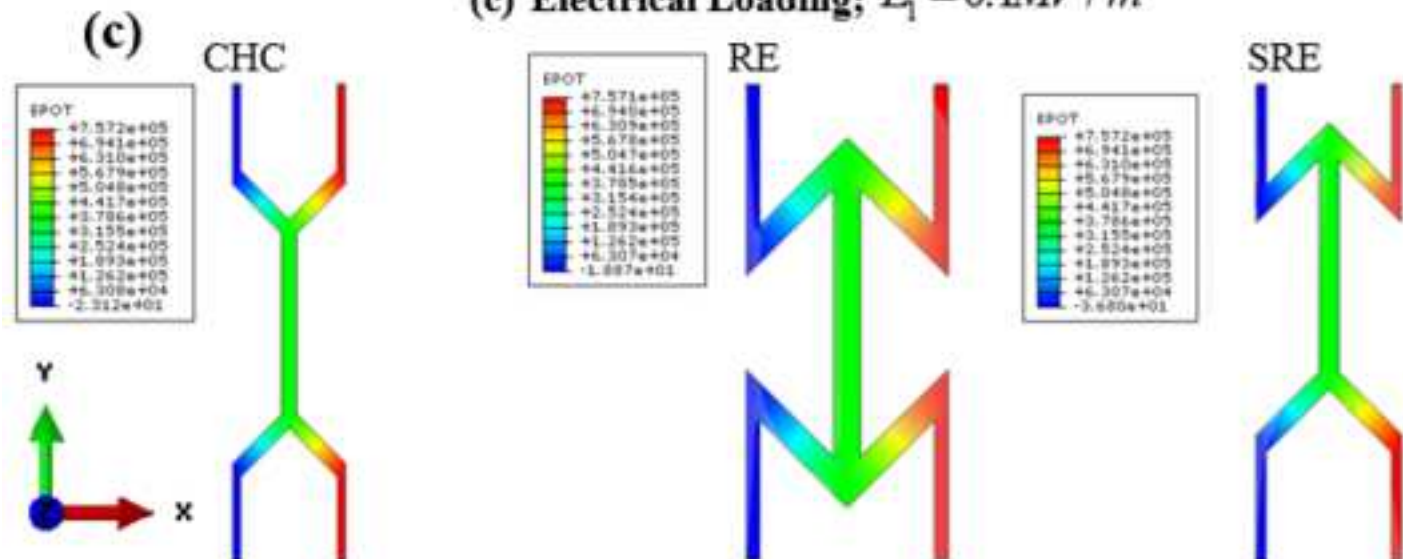


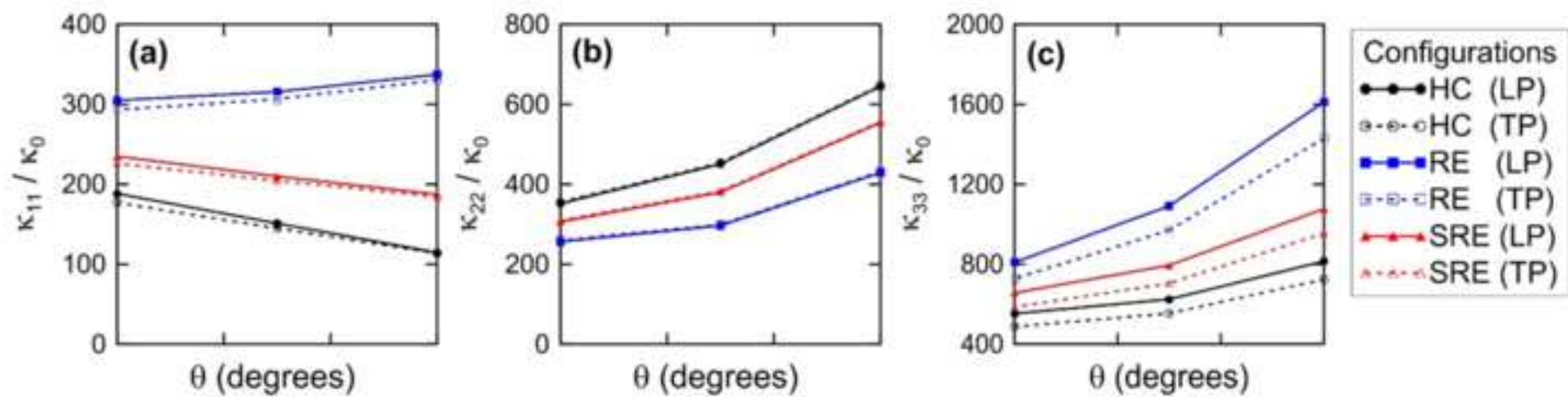


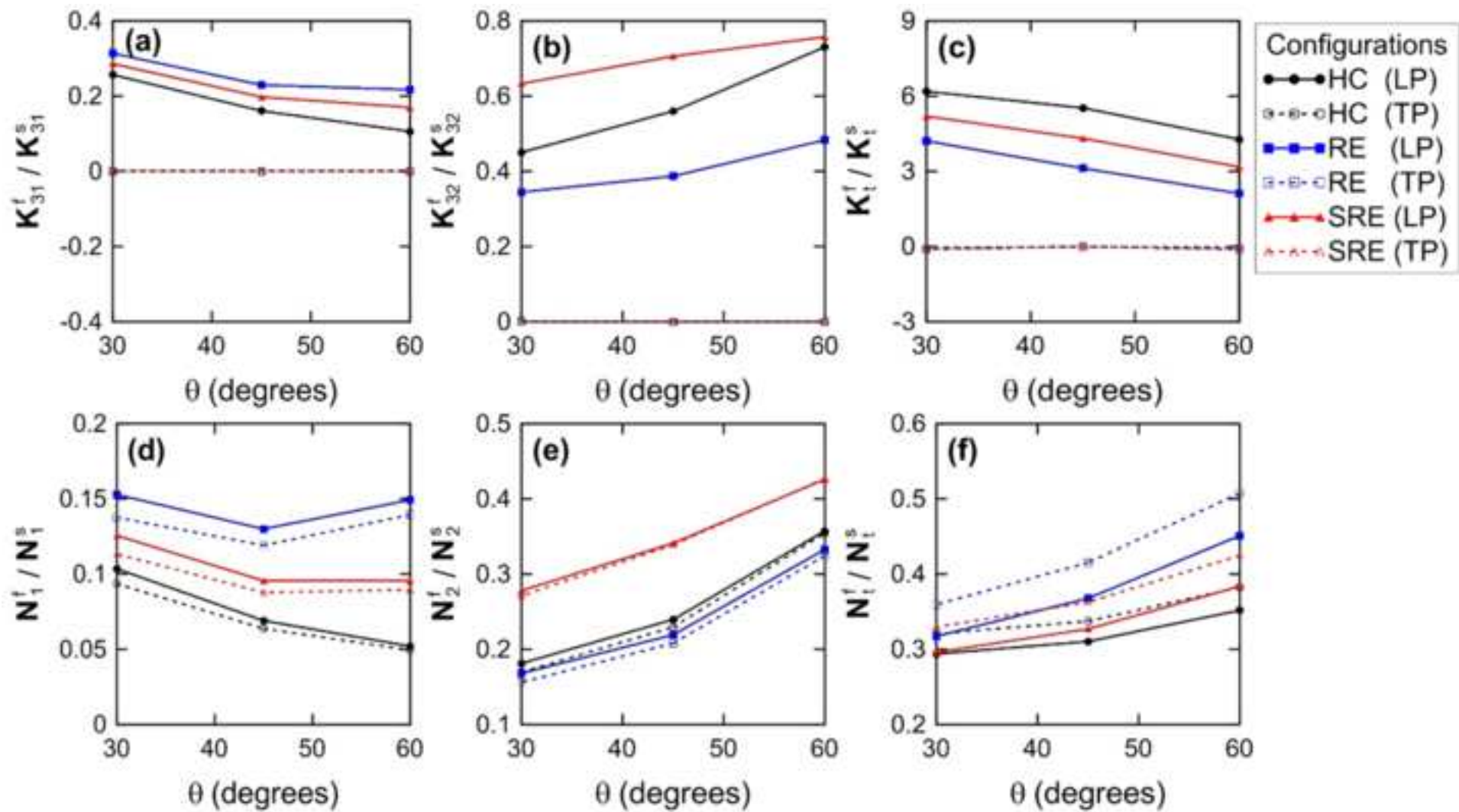
**(a)****(b)**





(a) Uniaxial Loading; $\varepsilon_{11} = 0.1\%$ **(b) Shear Loading; $\gamma_{12} = 0.1\%$** **(c) Electrical Loading; $E_1 = 0.1MV/m$** 





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

3

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

6

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

8

9 Figure 4: The overall elastic constants of piezoelectric HC cellular network at various angles (30°,
10 45°, 60°).

11

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

14

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

20

21 Figure 7: The overall piezoelectric constants of HC cellular network at various angles (30°, 45°,
22 60°).

23

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

25

26 Figure 9: Normalized FOMs of HC cellular network at various angles (30°, 45°, 60°). (a)

27 hydrostatic strain coefficient, (b) hydrostatic figure of merit, (c) acoustic impedance (d) $Z-d_h \cdot g_h$

28 relation

29

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

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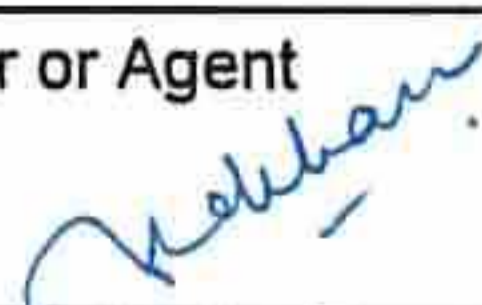
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Taryn Dollings
Editorial Coordinator
Materials Research Bulletin

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 auxetic 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 Bendsoe and Kikuchi [3] to design 1-3 piezocomposites with optimal performance characteristics for hydrophone applications. When design for maximum d_h and $d_h \cdot g_h$, 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 $d_h \cdot g_h$ 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 in 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 $d_h \cdot g_h$ 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 $d_h \cdot g_h$ 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 Poursolhjouy [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 explain 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 follow 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 condition 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.

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- [15] S. Li, Boundary conditions for unit cells from periodic microstructures and their implications, *Compos. Sci. Technol.* 68 (2008) 1962–1974.

Piezoelectric metamaterial with negative and zero Poisson's ratios

Khan, K. A.

2019-09-28

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