Universal converse flexoelectricity in dielectric materials via varying electric field direction

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ABSTRACT

Flexoelectricity is a symmetry independent electromechanical coupling phenomenon that outperforms piezoelectricity at micro and nanoscales due to its size-dependent behavior arising from gradient terms in its constitutive relations. However, due to this gradient term flexoelectricity, to exhibit itself, requires specially designed geometry or material composition of the dielectric material. First of its kind, the present study put forward a novel strategy of achieving electric field gradient and thereby converse flexoelectricity, independent of geometry and material composition of the material. The spatial variation of the electric field is established inside the dielectric material, Ba$_{0.67}$Sr$_{0.33}$TiO$_3$ (BST), by manipulating electrical boundary conditions. Three unique patterns of electrode placement are suggested to achieve this spatial variation. This varying direction of electric field gives rise to electric field gradient, the prerequisite of converse flexoelectricity. A multi-physics coupling based theoretical framework is established to solve the flexoelectric actuation by employing isogeometric analysis (IGA). Electromechanically coupled equations of flexoelectricity are solved to obtain the electric field distribution and the resulting displacements thereby. The maximum displacements of 0.2 nm and 2.36 nm are obtained with patterns I and II, respectively, while pattern III can yield up to 85 nm of maximum displacement.

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1. Introduction

Flexoelectricity has gained increasing interest from researchers in recent times as a symmetry independent physical phenomenon exhibiting electromechanical coupling in dielectric materials. A linear relationship between mechanical strain and induced electrical polarization is defined as direct flexoelectricity, whereas the phenomenon of generation of mechanical strain in linear proportionality to the gradient of the electric field is termed as converse flexoelectricity. Due to gradient terms in its constitutive relationships, flexoelectricity shows a strong size-dependent behavior. Researchers have used flexoelectric response in micro and nano electromechanical systems (MEMS/NEMS) where the piezoelectric response is of little significance. Kogan et al. [1] presented the first phenomenological report on a deformation gradient-based electromechanical coupling. A rough estimate of flexoelectric coefficients was provided, but it was termed as a type of piezoelectricity only. Mindlin [2] extended the linear theory of piezoelectricity by accounting for the contribution of polarization gradient in the stored energy function. Mindlin showcased the existence of electromechanical coupling even in centrosymmetric materials. Since then, an increasing amount of effort and time has been spent in investigating the physical behavior of flexoelectric materials, both theoretically and experimentally. Tagantsev [3] was the first to separately represent piezoelectric and flexoelectric effects in crystalline dielectrics. However, for several decades of its first evidence in solids, flexoelectricity was given a little attention, primarily because of its weak response. It was only after systematic experimental works conducted by the group of Cross [4–8] in the early 2000’s that the flexoelectricity was revived as an alternative of piezoelectricity at micro and nano scales. Theoretical evaluation of gradient elasticity-based electrotechnical response in dielectric materials has been a subject of interest for the research community even before these experimental works. Flexoelectric effect is a gradient-induced phenomenon, i.e., it requires inhomogeneous mechanical strain (for direct effect) or inhomogeneous electric field (for converse effect) to generate a net output. This inhomogeneity can be induced in different ways. One of the most common ways employed to get the required gradient is by having the specially designed geometry of the sample. Truncated pyramid shape was employed for the computational evaluation of flexoelectric characteristics of dielectric solids by Abdollahi et al. [9]. Smooth meshfree basis functions were employed to deal with the higher-order electromechanical coupling of flexoelectricity. Later, Abdollahi et al. [10] reinvestigated the evaluation of flexoelectric coefficients by compression of the truncated pyramid to address the issue of overestimation of properties by this popular method. The order of overestimation of the properties was systematically established as a function of pyramid configuration. Qi et al. [11] presented a unique design by generating a wavy piezo ribbons embedded silicon rubber-based micro structure for improvement in energy harvesting. The curved structure of piezoelectric ribbons was used as the source of inhomogeneous deformations resulting in a flexoelectric response. Qi et al. [12] numerically investigated a curved flexoelectric microbeam for direct and converse flexoelectric effect. Several other studies [13–21] have been reported using special geometry as a source of nonuniform deformation field to induce flexoelectricity. Another way extensively exploited for attaining the prerequisite of flexoelectricity, i.e., the gradient of strain, is by having appropriate boundary and loading conditions. For this purpose, cantilever-shaped energy harvesters and sensors, due to the presence of nonuniform strain in bending, have been
Vastly investigated in the past [22–33], variable mechanical characteristics can result in varying strain distribution within the domain. A few studies have suggested the use of special composite materials to achieve variation in mechanical properties and thereby a strain gradient [34–39]. This method eliminates the need for special designs and dimensions of the devices to be employed, however, it requires an additional effort at manufacturing stage for compositional variation. The variety of computational techniques and experimental efforts observed from the literature focus mainly on investigating, optimizing, and improving the direct flexoelectric effect while studies on the converse flexoelectric effect are comparatively less. Fu et al. [40] experimentally evaluated the converse flexoelectric coefficients in a highly electrically susceptible $\text{Ba}_{0.67}\text{Sr}_{0.33}\text{TiO}_3$ (BST) trapezoidal block. The resulting $\mu_{11}$ was found to be in good agreement with that measured by direct effect in their previous studies. Abdollahi et al. [9] computationally evaluated the direct and converse flexoelectricity in a trapezoidal block in a two-dimensional (2D) domain. Experimental analysis of converse flexoelectric effect in a $\text{SrTiO}_3$ crystal was performed by Zalesskii and Rumyantseva [41], under an inhomogeneous strain produced in bending and varying temperature. In a comparative study of piezoelectricity and flexoelectricity, Abdollahi and Arias [42] presented the potentials of piezoelectric and flexoelectric electromechanical coupling phenomena, in both direct and converse effects. The size dependence of net output from both phenomena was compared and flexoelectricity was found to be more effective at smaller scales. A mixed finite elements-based FEA formulation was developed by Mao et al. [43], for direct and converse flexoelectric effects while considering the electromechanical coupling of polarization with both symmetrical and rotational strain gradients. Additional nodal degrees of freedom were introduced to reduce the $C^1$ continuous problem to a $C^0$ continuous problem. An actuation method employing a biconcave curved beam of polyvinylidene fluoride (PVDF), using the finite element method was presented by Wu et al. [44]. Converse flexoelectric effect has also been utilized in vibration control application by Fan and Tzou [45] as an actuator mechanism. A cantilever beam laminated with a flexoelectric layer was used to demonstrate the influence of actuating effect of the flexoelectric layer. Very recently, Liu et al. [46] presented a theoretical and experimental study on direct and converse flexoelectric effects. A mixed finite element method for numerical simulations was used and piezoresponse force microscopy (PFM) was used in experimental studies.

Upon reviewing the existing literature, it is revealed that although a fair amount of attention is being given to the computational evaluation of direct flexoelectricity, there exists a lack of focus on converse flexoelectricity as compared to direct effect. Moreover, the previous attempts on studying the converse flexoelectric effect use engineered structures or composition as a source of inhomogeneity to induce the effect. This method, although used widely, puts a constraint on the choice of geometry and/or material composition for the use in relevant applications. In this study, we report an approach for generating converse flexoelectricity in a regular shaped homogeneous dielectric material by spatial variation of electric field direction. The present method incorporates a spatial variation of the electric field vector in the material by manipulating electrical boundary conditions. This is done by employing different configurations of electrode placement. A multi-physics coupled field model is developed and solved using isogeometric analysis (IGA) on a 2D material sample of BST. The switching of a polarization vector field in a varying fashion occurs due to the nonconventional electrodoping, which is
captured by solving electrostatic equilibrium. The resulting actuation is computed by employing coupled flexoelectric equations. Three different patterns of varying electric field vector are attained by changing the location of application of electric potential and grounding on the sample. The proposed method provides a relatively easy to implement a way of inducing converse flexoelectric effect for micro- and nano-scale applications.

2. Methodology

A variety of methods can be utilized to generate a varying electric fields in order to achieve converse flexoelectricity in dielectric materials. Figure 1(a,b) shows the methods that have been utilized in the past for this purpose, namely, geometry variation and material composition variation. In Figure 1(c), the variation of electric field direction is demonstrated schematically to achieve electric field gradient. To demonstrate the converse flexoelectric effect generated due to spatial variation of electric field direction, a 2D rectangular geometry is selected for computation. Assumptions of plane strain elasticity and electrostatics in 2D are employed while formulating the equations used in IGA. The multi-physics computational framework required for the analysis of the converse flexoelectric effect will be developed using flexoelectric material constitutive law. The IGA formulation is developed by keeping these equations as the basis. The detailed formulation is discussed in subsequent sections.

2.1. Flexoelectric material law

Flexoelectricity is described by a symmetry independent and size-dependent correlation between electrical and mechanical field variables. Linear proportionality between polarization and strain gradient is termed as direct flexoelectricity whereas the linear proportionality of mechanical stress with electric field gradient is termed as converse flexoelectricity. In this section, we will derive this correlation and later based on the constitutive equations of flexoelectricity, an isogeometric numerical model will be developed. The electrical Gibbs free energy density of a dielectric material having piezoelectric and flexoelectric effects can be written as [9],

$$H = \frac{1}{2} C_{ijkl} S_{ij} S_{kl} - e_{ijkl} E_{i} S_{kl} + f_{ijkl} E_{i} S_{jk,l} + d_{ijkl} E_{i} E_{j} S_{kl} - \frac{1}{2} \varepsilon_{ij} E_{i} E_{j}$$

(1)

Figure 1. Schematic of arrangements to induce converse flexoelectricity, (a) geometry-dependent electric field gradient, (b) varying material composition for electric field gradient, and (c) varying direction of electric field for generating electric field gradient.
where $E$ and $S$ are the electric field vector and strain tensor, respectively. $C, f, d,$ and $\mu$ denote fourth-order material elastic stiffness tensor, fourth-order direct flexoelectric tensor, converse flexoelectric tensor, and second-order dielectric tensor, respectively. Subscripts $i, j, k,$ and $l$ are the representatives of orthogonal cartesian coordinate directions. The first term denotes the contribution of mechanical strain energy, while the second term is the contribution of piezoelectricity. Third and fourth terms are the direct and converse flexoelectricity contributions to the energy density function. The last term is the electrostatic contribution arising from applied potential. It has been shown that the terms pertaining to direct and converse flexoelectric contributions can be represented by a single material tensor in the following form [15],

$$H = \frac{1}{2} C_{ijkl} S_{ij} S_{kl} - e_{ijkl} E_i S_{kl} - \mu_{ijkl} E_i S_{jk, l} - \frac{1}{2} \varepsilon_{ij} E_j E_k$$

(2)

Here $\mu_{ijkl} = d_{ijk} - f_{ijkl}$ is the fourth-order flexoelectric coefficients’ tensor. We write stress and electric displacement as,

$$\hat{T}_{ij} = \frac{\partial H}{\partial S_{kl}} = C_{ijkl} S_{ij} - e_{ijkl} E_k$$

(3)

and

$$\hat{D}_i = - \frac{\partial H}{\partial E_j} = e_{ijkl} S_{kl} + \varepsilon_{ij} E_j + \mu_{ijkl} S_{jk, l}$$

(4)

Next, the higher-order stress (moment stress or hyper stress) and higher-order electric displacement (electric quadrupole) are defined as,

$$\tilde{T}_{ijk} = \frac{\partial H}{\partial S_{jk, l}} = -\mu_{ijkl} E_i$$

(5)

and

$$\tilde{D}_{ij} = - \frac{\partial H}{\partial E_{i, j}} = 0$$

(6)

The physical stress can be written as,

$$T_{ij} = \hat{T}_{ij} - \tilde{T}_{ijk, k} = C_{ijkl} S_{ij} - e_{ijkl} E_k + \mu_{ijkl} E_{i, k}$$

(7)

Similarly, the physical electric displacement can be written as,

$$D_i = \hat{D}_i - \tilde{D}_{ij, j} = e_{ijkl} S_{kl} + \varepsilon_{ij} E_j + \mu_{ijkl} S_{jk, l}$$

(8)

For non-piezoelectric material, the piezoelectric terms in Equations (7) and (8) can be omitted. Rewriting Equations (7) and (8), the constitutive equations of flexoelectric effect can be written as,

$$D_i = \varepsilon_{ij} E_j + \mu_{ijkl} S_{jk, l}$$

(9)

and

$$T_{ij} = C_{ijkl} S_{ij} + \mu_{ijkl} E_{i, j}.$$  

(10)
Equations (9) and (10) are the constitutive equations for direct and converse flexoelectric effects, respectively. The electrical Gibbs free energy, can be written as,

\[ H = \frac{1}{2} \int_{\Omega} \left( \tilde{T}_{ij} S_{ij} + \tilde{T}_{ijk} S_{ij} \right) d\Omega - \int_{\Gamma} t_i q_i ds + \int_{\Gamma} Q \phi ds \]  

(11)

Here the last two terms are the work potentials of traction \( t_i \) and surface charge density \( Q \).

2.2. Isogeometric analysis

2.2.1. Discretization, field variables, and derivatives

Isogeometric analysis, in contrast to finite element analysis (FEA), uses higher-order nonuniform rational B-spline (NURBS) as basis functions (Figure 2(c)), integrating CAD and CAE. The need for using IGA in the analysis of flexoelectricity arises due to higher-order coupling between primary variables in constitutive law (Equations (11)–(12)). To evaluate the gradient of strain and electric field, basis functions should be at least \( C^1 \) continuous, which is enabled by the NURBS basis functions of IGA. Control points used to generate NURBS are the points used for discretization of geometry and assigning degrees of freedom. Two mechanical and one electrical degrees of freedom at each control point are assigned. The control points trace the envelop of the surface that they are used to generate, but do not necessarily lie on the surface itself. A typical example of

\[ \xi_1 = [0, 0, 0, 1, 2, 3, 3, 3] \text{ and } \xi_2 = [0, 0, 1, 1], \]

Figure 2. Isogeometric discretization, (a) Index space for a domain defined by knot vectors \( \eta_1 \) and \( \eta_2 \) \( \xi_1 \) \( \xi_2 \) \( \xi_3 \) \( \xi_4 \) \( \xi_5 \) \( \xi_6 \) \( \xi_7 \) \( \xi_8 \) \( \eta_1 \) \( \eta_2 \) \( \eta_3 \) \( \eta_4 \), (b) parametric space before normalization, and normalized parametric space, (c) associated NURBS basis functions.
discretization in IGA is depicted in Figure 2(a,b), wherein the index space associated with the particular knot vectors and creation of parametric space is demonstrated. The geometry of the 2D domain is represented using NURBS basis functions as,

\[ X^* = \sum_{i=1}^{n_x} \sum_{j=1}^{n_y} R_{ij}^{p_r,p_s}(r,s)X_{ij} \]  

(12)

where \( X = \{x, y\} \) is the location of a control point. The number of control points in \( x \) and \( y \) directions are denoted by \( n_x \) and \( n_y \), respectively. The order of polynomial in \( r \) and \( s \) directions are given by superscripts \( p_r \) and \( p_s \), respectively. Using the same NURBS basis functions to denote displacement field, \( q = \{u, v\} \), at a control point,

\[ q^*(x, y) = \sum_{i=1}^{n_x} \sum_{j=1}^{n_y} R_{ij}^{p_r,p_s}(r,s)q_{ij} \]  

(13)

Similarly, the electric potential at a point, \( \phi^* \) is approximated as,

\[ \phi^*(x, y) = \sum_{i=1}^{n_x} \sum_{j=1}^{n_y} R_{ij}^{p_r,p_s}(r,s)\phi_{ij} \]  

(14)

The NURBS basis used in equations (21)–(23), at a control point with indices \( i \) and \( j \) and associated weight \( w \), is represented as,

\[ R_{ij}^{p_r,p_s}(r,s) = \frac{N_{i,p_r}(s)N_{j,p_s}(r)w_{ij}}{\sum_{i=1}^{n_x} \sum_{j=1}^{n_y} N_{i,p_r}(s)N_{j,p_s}(r)w_{ij}} \]  

(15)

where \( N \) is the B-spline basis function. Equations (12)–(14) denote the field variables in terms of spatially varying NURBS basis functions and values of field variables at control points. Thus, while deriving the quantities such as strain and electric field and their gradients, one needs to differentiate NURBS basis functions with respect to spatial coordinates, which is facilitated by higher-order continuity involved in these basis functions. Writing the strain field in terms of displacements,

\[ \{S\} = \frac{\partial \{q^*\}}{\partial \{X\}} \]  

(16)

Replacing \( \{q\} \) from Equation (13) into Equation (16),

\[ \{S\} = \frac{\partial \{R\}}{\partial \{X\}} \{q\} \]  

(17)

or,

\[ \begin{bmatrix} S_{11} \\ S_{22} \\ S_{12} \end{bmatrix} = \begin{bmatrix} \frac{\partial R_{11}}{\partial x} & 0 & \frac{\partial R_{12}}{\partial x} & 0 & \ldots & \frac{\partial R_{1,p}}{\partial x} & 0 \\ 0 & \frac{\partial R_{11}}{\partial y} & 0 & \frac{\partial R_{12}}{\partial y} & \ldots & 0 & \frac{\partial R_{1,p}}{\partial y} \\ \frac{\partial R_{21}}{\partial x} & \frac{\partial R_{22}}{\partial x} & \frac{\partial R_{22}}{\partial y} & \ldots & \frac{\partial R_{2,p}}{\partial x} & \frac{\partial R_{2,p}}{\partial y} \\ \end{bmatrix} \begin{bmatrix} u_1 \\ v_1 \\ u_2 \\ \vdots \\ v_2 \\ \vdots \\ v_{n_y} \end{bmatrix} \]  

(18)

or,
\{ S \} = [B_q] \{ q^p \} \tag{19}

where \( n_p \) is the total number of control points, i.e. \( n_p = n_x \times n_y \) and \([B_q]\) is the first order derivatives’ matrix of NURBS basis functions. Next, we evaluate strain gradient by differentiating Equation (19) with respect to spatial coordinates,

\[ \frac{\partial \{ S \}}{\partial \{ X \}} = [H_q] \{ q \} \tag{20} \]

Here \([H_q]\) denotes the second-order derivatives’ matrix of NURBS basis functions (corresponding to strain gradient), known as Hessian matrix. (21)

\[
[H_q] = \begin{bmatrix}
\frac{\partial^2 R_1}{\partial x^2} & 0 & \frac{\partial^2 R_2}{\partial x^2} & 0 & \frac{\partial^2 R_3}{\partial x^2} & 0 & \cdots & \frac{\partial^2 R_{np}}{\partial x^2} & 0 \\
0 & \frac{\partial^2 R_1}{\partial y^2} & 0 & \frac{\partial^2 R_2}{\partial y^2} & 0 & \frac{\partial^2 R_3}{\partial y^2} & \cdots & 0 & \frac{\partial^2 R_{np}}{\partial y^2} \\
\frac{\partial^2 R_1}{\partial x \partial y} & 0 & \frac{\partial^2 R_2}{\partial x \partial y} & 0 & \frac{\partial^2 R_3}{\partial x \partial y} & 0 & \cdots & 0 & \frac{\partial^2 R_{np}}{\partial x \partial y} \\
0 & \frac{\partial^2 R_1}{\partial x \partial y} & 0 & \frac{\partial^2 R_2}{\partial x \partial y} & 0 & \frac{\partial^2 R_3}{\partial x \partial y} & \cdots & 0 & \frac{\partial^2 R_{np}}{\partial x \partial y} \\
\frac{\partial^2 R_1}{\partial y^2} & \frac{\partial^2 R_2}{\partial y^2} & \frac{\partial^2 R_3}{\partial y^2} & \cdots & \frac{\partial^2 R_{np}}{\partial y^2} \\
\end{bmatrix} \tag{21}
\]

In the similar way, an electric field can be related to electric potential as,

\[ \{ E \} = -\frac{\partial \{ \phi \}}{\partial \{ X \}} \tag{22} \]

or,

\[
\begin{bmatrix}
E_x \\
E_y
\end{bmatrix} = - \begin{bmatrix}
\frac{\partial R_1}{\partial x} & \frac{\partial R_2}{\partial x} & \frac{\partial R_3}{\partial x} & \cdots & \frac{\partial R_{np}}{\partial x} \\
\frac{\partial R_1}{\partial y} & \frac{\partial R_2}{\partial y} & \frac{\partial R_3}{\partial y} & \cdots & \frac{\partial R_{np}}{\partial y}
\end{bmatrix}
\begin{bmatrix}
\phi_1 \\
\phi_2 \\
\vdots \\
\phi_{np}
\end{bmatrix} \tag{23}
\]

or,

\[ \{ E \} = - [B_\phi] \{ \phi \} \tag{24} \]

As required by the governing law of flexoelectricity, we now evaluate the derivative of the electric field. Differentiating Equation (24) with respect to spatial coordinates,

\[ \frac{\partial \{ E \}}{\partial \{ X \}} = \begin{bmatrix}
\frac{\partial E_x}{\partial x} \\
\frac{\partial E_x}{\partial y} \\
\frac{\partial E_y}{\partial x} \\
\frac{\partial E_y}{\partial y}
\end{bmatrix} \tag{25} \]

or,
\[
\begin{align*}
\begin{bmatrix}
\frac{\partial E_x}{\partial x} & \frac{\partial E_y}{\partial y} & \frac{\partial E_z}{\partial z} \\
\frac{\partial E_x}{\partial y} & \frac{\partial E_y}{\partial y} & \frac{\partial E_z}{\partial y} \\
\frac{\partial E_x}{\partial z} & \frac{\partial E_y}{\partial z} & \frac{\partial E_z}{\partial z}
\end{bmatrix}
& =
\begin{bmatrix}
\frac{\partial R_1}{\partial x} & 0 & 0 & \cdots & \frac{\partial R_p}{\partial x} & 0 \\
0 & \frac{\partial R_1}{\partial y} & 0 & \cdots & 0 & \frac{\partial R_p}{\partial y} \\
0 & 0 & \frac{\partial R_1}{\partial y} & \cdots & 0 & \frac{\partial R_p}{\partial y}
\end{bmatrix}
\begin{bmatrix}
E_{x1} \\
E_{y1} \\
E_{z1} \\
E_{x2} \\
E_{y2} \\
E_{z2} \\
\vdots \\
E_{xnp} \\
E_{ynp}
\end{bmatrix}
\end{align*}
\]

Replacing the values of the electric field in terms of \([B_\phi]\) and solving, Equation (26) becomes,

\[
\begin{align*}
\begin{bmatrix}
\frac{\partial E_x}{\partial x} & \frac{\partial E_y}{\partial y} & \frac{\partial E_z}{\partial z} \\
\frac{\partial E_x}{\partial y} & \frac{\partial E_y}{\partial y} & \frac{\partial E_z}{\partial y} \\
\frac{\partial E_x}{\partial z} & \frac{\partial E_y}{\partial z} & \frac{\partial E_z}{\partial z}
\end{bmatrix}
& =
\begin{bmatrix}
\frac{\partial^2 R_1}{\partial x^2} & \frac{\partial^2 R_1}{\partial x \partial y} & \frac{\partial^2 R_1}{\partial x \partial z} & \cdots & \frac{\partial^2 R_1}{\partial x \partial p} & \frac{\partial^2 R_1}{\partial y \partial p} \\
\frac{\partial^2 R_1}{\partial y \partial x} & \frac{\partial^2 R_1}{\partial y^2} & \frac{\partial^2 R_1}{\partial y \partial z} & \cdots & \frac{\partial^2 R_1}{\partial y \partial p} & \frac{\partial^2 R_1}{\partial z \partial p} \\
\frac{\partial^2 R_1}{\partial z \partial x} & \frac{\partial^2 R_1}{\partial z \partial y} & \frac{\partial^2 R_1}{\partial z^2} & \cdots & \frac{\partial^2 R_1}{\partial z \partial p} & \frac{\partial^2 R_1}{\partial p \partial p}
\end{bmatrix}
\begin{bmatrix}
\phi_1 \\
\phi_2 \\
\phi_3 \\
\phi_4 \\
\vdots \\
\phi_{np-1} \\
\phi_{np}
\end{bmatrix}
\end{align*}
\]

or,

\[
\frac{\partial \{E\}}{\partial \{X\}} = -[H_\phi] \{\phi\}
\]

where \([H_\phi]\) is the Hessian of the NURBS basis functions corresponding to electric field gradient.

### 2.3. Governing equations

In this subsection, we derive the equilibrium equations of the system using constitutive material law of flexoelectricity given by Equations (9) and (10). Using the matrix notations, the Equation (11) can be rewritten as,

\[
H = \frac{1}{2\Omega} \left( \int_T \{S\}^T \{T\} - \{\phi\}^T \{\phi\} \right)
+ \left[ \{E\}^T \{\mu\} \right]
- \int_{\Gamma_r} \{q^s\}^T \{t\} ds + \int_{\Gamma^p} \{\phi\}^T \{Q\} ds
\]

It is to be noted that \(T\) in superscripts denotes the transpose of a matrix and not the usual notation of the stress vector. Further, replacing the values of stress, strain, strain gradient, electric field, and electric field gradient in Equation (29),

\[
H = \frac{1}{2\Omega} \left( \{q\}^T [B]^T[C][B] \{q\} + \{q\}^T [H_q]^T [\mu]^T [B_\phi] \{\phi\} 
- \{\phi\}^T [B_\phi] \{\phi\}^T [\mu] [H_q] \{\phi\} \right)
+ \int_{\Gamma_r} \{q\}^T \{R\}^T \{t\} ds + \int_{\Gamma^p} \{\phi\}^T \{R\}^T \{Q\} ds,
\]

or,
\[
H = \frac{1}{2} \left( \begin{pmatrix} q \end{pmatrix}^T \left[ K_{qq} \right] \begin{pmatrix} q \end{pmatrix} + \begin{pmatrix} q \end{pmatrix}^T \left[ K_{q\phi} \right] \begin{pmatrix} \phi \end{pmatrix} - \begin{pmatrix} \phi \end{pmatrix}^T \left[ K_{\phi q} \right] \begin{pmatrix} q \end{pmatrix} \right) - \begin{pmatrix} q \end{pmatrix}^T \begin{pmatrix} R \end{pmatrix}^T \begin{pmatrix} t \end{pmatrix} + \begin{pmatrix} \phi \end{pmatrix}^T \begin{pmatrix} R \end{pmatrix}^T \begin{pmatrix} Q \end{pmatrix} \right)
\]

(31)

Equation (31) is dependent on two field variables, \( \{ q \} \) and \( \{ \phi \} \). To derive the equilibrium equation in mechanical and electrical domains, energy expression given by (31) is minimized for \( \{ q \} \) and \( \{ \phi \} \) as,

\[
\frac{\partial H}{\partial q} = [K_{qq}] \{ q \} + [K_{q\phi}] \{ \phi \} - \{ F_m \} = 0
\]

(32)

and,

\[
\frac{\partial H}{\partial \phi} = -[K_{\phi q}] \{ q \} + [K_{\phi \phi}] \{ \phi \} + \{ F_Q \} = 0
\]

(33)

Simplifying Equations (32) and (33), we get the governing equations of direct and converse effects as,

\[
[K_{\phi q}] \{ \phi \} - [K_{q\phi}] \{ q \} = \{ F_Q \}
\]

(34)

\[
[K_{qq}] \{ q \} + [K_{q\phi}] \{ \phi \} = \{ F_m \}
\]

(35)

Matrices \([K_{qq}],[K_{q\phi}],\) and \([K_{q\phi}]\) are the mechanical, electrical, and electromechanical coupling stiffness matrices. \(\{F_m\}\) and \(\{F_Q\}\) are the mechanical and electrical force vectors. The details of the material matrices, stiffness matrices, and force vectors are given in Appendix A. Equations (34) and (35) are solved in a coupled sense for computing the potential distribution in the domain and displacement field due to this potential.

3. Numerical simulations

The theoretical formulation discussed in section 2 is employed for analyzing the converse flexoelectric effect generated due to varying electric field direction inside a dielectric material. Three different combinations of electrical boundary conditions are discussed to achieve different patterns of spatially varying electric field within the material. Variation in electric field direction generates a gradient of the electric field in \( x \) and \( y \) directions, resulting in a net displacement in the material.

3.1. Verification of IGA model

Before proceeding to the main studies of the present work, we have verified the formulation developed in section 2, against some benchmark problems available in the literature.

First, we take an example solved in reference [9]. A cantilever beam subjected to a point load at its free end is investigated for its flexoelectric coupling coefficients. Figure 3(a) depicts the comparison of the present results and those of the reference results. The values obtained from the present formulation can be seen to be in good agreement with those of the reference. Further, another example from the same reference
is replicated to calculate the electric field distribution across the thickness of the beam due to flexoelectric coupling (Figure 3(b)).

Next, a trapezoid-shaped sample with a base width of 2250 µm, top width of 750 µm, and height of 750 µm is subjected to 6 MPa pressure on the top surface. The bottom surface is fixed and the top surface is electrically grounded. The color contours of strain and electric potential generated inside the sample are plotted in Figure 3(c, Figure 3d). An excellent match with the reference [9] results is observed. It is worth mentioning here that the validations presented in Figure 3(a–d) consider the coupling of direct and converse flexoelectric effect. Thus, these validations are sufficient to prove the validity of the computational model in evaluating flexoelectricity in its entirety, i.e. direct and converse flexoelectric effects.

3.2. Pattern I

As mentioned earlier, three different patterns are proposed with the varying direction of the electric field, obtained by applying electric potential and ground to selected portions of the computational domain. To demonstrate the effectiveness of the proposed method in generating converse flexoelectricity, independent of geometry and composition, a square-shaped 2D block of BST is selected. The side of the square is 50 µm and it is fixed at the bottom surface. The material properties of BST used are listed in Table 1. Unless otherwise stated, the same geometrical and material properties are used for all the studies conducted in this paper hereafter. Usually, when a dielectric material is subjected to electrical loading in order to work as an actuator, the top and bottom surfaces are covered with the electrodes and an electric field gradient is achieved due to varying
material composition or cross-section area. However, in this case, we use a homogeneous material with a uniform cross section, while for generating an inhomogeneous electric field, electrode placement is changed from the conventional arrangement.

In the first case, one of the electrodes is placed at the right half of the top surface while another at the left half of the bottom surface, as depicted in Figure 5(a). This arrangement is referred to as pattern I. Bottom electrode is applied with an electric potential of 100 V while the top electrode is grounded. Electric field lines have an inherent nature to flow from lower potential to higher potential in a medium. Due to this reason, the majority of electric field lines tend to move from the lower electrode to the upper electrode diagonally, as shown in Figure 5(b). The magnitude of the electric field is shown by a color map whereas the electric field direction at control points is depicted by arrows. Extreme values of electric field intensity are noticed near the tips of both the electrodes. This can be credited to a sudden change in electric potential from electrode surface to surrounding dielectric material. In contrast to the usual arrangement, which generates a constant electric field and no electric field gradient, the pattern I having electrodes at top and bottom surfaces partially, gives an inhomogeneous electric field. This inhomogeneous electric field is the source of large converse flexoelectricity and hence a large displacement due to an applied electric field, witnessed in the BST sample being studied. These large displacements are generally localized in nature and are not appropriate for practical applications. Thus, to demonstrate the actuation in a more practical sense, a constant but unknown displacement has been prescribed on the top surface, i.e. the target surface for actuation.

It is to be mentioned that before proceeding to numerical simulations, we have performed a convergence mesh study to establish the reliability of the computational model.

| Parameter                        | Value | Reference |
|----------------------------------|-------|-----------|
| Young’s modulus, $E$ (GPa)       | 153   | [47]      |
| Poisson’s ratio, $\nu$           | 0.33  | [47]      |
| Flexoelectric coefficient, $\mu_{12}$ ($\mu C/m$) | 8.5   | [13]      |
| Flexoelectric coefficient, $\mu_{11}$ ($\mu C/m$) | 100   | [13]      |
| Relative permittivity, $\varepsilon_r$ | 4100  | [13]      |

**Table 1.** Material parameters used for numerical simulations.

![Figure 4. Variation of displacement at the top surface with total number of degrees of freedom of the computational model.](image)
model. Figure 4 shows the mesh convergence results for the pattern I (Figure 5(a)). A good convergence is an observer at 326 degrees of freedom.

The linear proportionality of electric field gradient and resulting mechanical stress (Equation (10)) yields the deformation of the dielectric material. However, electric field gradient, in the case of the 2D domain has four unique components, namely, variations of $X$ component of the electric field in $X$ and $Y$ direction; and variation of $Y$ component of the electric field in $X$ and $Y$ directions. The variation of horizontal displacement along length at mid height (path AA’) of the structure is depicted by the right y-axis of Figure 6, where X-displacement gradually reduces toward mid-span. It is observed that the converse flexo-electric effect generates displacements that are significantly low scale (nanometers), however, technologically relevant for nano and micro scales and precision engineering applications. Due to high variation in electric field direction near the mid of path AA’ (Figure 5), the highest values of displacement are observed here. As for the
Y-displacement, which is plotted on the left y-axis of Figure 6 at mid span (path BB’) from bottom surface to top, it has an extreme positive value near top and bottom ends while at the extremes it has a very small value with a sudden jump following. This is due to the mechanical constraints applied at the top and bottom surfaces of the structure.

The simulations presented till now are performed on a square-shaped sample \((a = b)\) and electrode length \(L = a/2\). However, the magnitude and direction of the electric field are highly dependent on the placement of electrodes and the aspect ratio of the structure. To inspect the effect of aspect ratio \((R = a/b)\) and electrode length ratio \((r = L/b)\) on the actuation of the material, variations of horizontal and vertical displacements are plotted with variations in aspect ratio and electrode length ratio in Figure 7(a, b), respectively. It is to be mentioned that Figure 7(a) is plotted with a constant value of \(r = 0.5\) while Figure 7(b) is plotted assuming a constant value of \(R = 0.5\). With decreasing aspect ratio, electric field gradient tends to become sharp due to less distance available to accommodate the high difference in minimum and maximum magnitudes. This leads to an increase in resulting displacement at the target surface, as observed in Figure 7(a). On the other hand, increasing the electrode length ratio increases the value of the displacements. Although the smaller values of \(r\), i.e. a small value of electrode length placed at opposite ends of the diagonal of a rectangle, have high values of electric field gradients, the active length of the target surface being actuated decrease. For a value of \(r = 1\) electric field may seem to have has a constant value and direction (vertically upward) in the entire computational domain, yielding zero electric field gradient. However, this is not true in the case of materials with flexoelectric coupling. The direct and converse flexoelectric effects being the manifestations of a single electromechanical coupling, as argued in references [9,48], tend to influence one another by changing boundary conditions. This can be also be understood by inspecting the governing equations (Equation (34) and (35)) of the flexoelectricity, where the coupling stiffness matrix used in the converse flexoelectric effect is the same as that in direct effect. Thus, the pattern I can generate high displacements locally, but fails to surpass the performance of the conventional pattern of electroding (top and bottom surface fully covered by electrodes) in actuating the entire target surface.

![Figure 7](image-url)

Figure 7. Variations of maximum X and Y displacements with (a) varying aspect ratio and (b) varying electrode length ratio.
3.3. Pattern II

Pattern I of electric field distribution was achieved by placing electrodes on the two opposite faces of the material. Pattern I creates an electric field gradient which is beneficial for increased flexoelectric actuation but at the same time also suffers from the disadvantage of the reduced total length of electrodes. Pattern II is achieved by extending the electrodes of the pattern I to their respective adjacent orthogonal sides, as shown schematically in Figure 8(a). More specifically, the ground electrode is applied on the left half of the bottom face and bottom half of the left face, whereas the other electrode with electric potential is placed on the right half of the top face and upper half of the right face. This arrangement ensures a similar type of electric field distribution in $x$ and $y$ directions, without a reduction in total electrode surface area from the conventional electroding. Electrode lengths are given in terms of side lengths of

![Figure 8](image_url)

**Figure 8.** Pattern II of spatially varying electric field in BST sample, (a) electrode placement, dimensions and boundary conditions of the sample, (b) electric field distribution inside the sample.

![Figure 9](image_url)

**Figure 9.** Variations of $X$ and $Y$ displacements, along AA’ and BB’ respectively for pattern II.
the sample. Upon analyzing the displacements obtained from pattern II, X-displacement on the right surface of the structure is chosen to be the target actuation. Figure 8(b) depicts the electric field distribution in the material due to an applied voltage of 100 V. The magnitude of the electric field is denoted by the colormap while arrows are indicative of electric field direction at the control points. The magnitude of the electric field has extreme values at the tips of the electrodes, which is the same as pattern I. The resulting displacement in the x and y direction will be obtained due to the combined effect of the gradient of the electric field in both the directions.

To investigate the electro-mechanical coupling of pattern II of spatially varying electric field gradient due to converse flexoelectric effect, horizontal and vertical components of displacement are scrutinized. The right Y-axis of Figure 9 (b) represents the spatial variation of X-component of displacement along path AA', while variation of Y-component of displacement along BB' is represented at left Y-axis of the structure. The magnitudes of displacements are larger than those in the case of the pattern I. This is due to the fact that while pattern I depends on the varying direction of electric field for converse flexoelectric effect, pattern II also has an increased span of electrodes to its advantage.

The results so far presented for pattern II are obtained by keeping aspect ratio R and electrode length ratio r constant at 0.5. However, pattern II, similar to the pattern I, shows dependence on these two ratios. This dependence is inspected graphically in Figure 10(a, b) for R and r, respectively. Varying R from 0 to 1 while keeping r constant at 0.5, gives an increase in the actuation due to sharp gradients of the electric field. On the other hand, Figure 10(b) follows an increasing trend in the actuation but only up to $r = 0.5$. Beyond $r = 0.5$, the value of actuation suddenly drops, due to the reduction in electric field gradient dominating over the increase in electrode span. This trend is observed only between $r = 0.5$ and 0.6, beyond the value of actuation starts to rise again. It is to be mentioned here that although the increasing values of r might seem to give better performance in terms of actuation, the values of r equal or close to 1 do not have practical significance because of the possibility of zero gap between the ground and electrical potential.

![Figure 10](image-url). Variations of maximum X and Y displacements with (a) varying aspect ratio and (b) varying electrode length ratio, for pattern II.
Thus, being an extension of pattern I, pattern II exhibits similar behavior in terms of electric field variations and electric field gradients. However, the two-axis symmetry of electrical boundary conditions gives rise to some remarkable variations to the converse flexoelectric behavior of pattern II, as compared to the pattern I.

3.4. Pattern III

The two patterns discussed so far were designed by altering the conventional arrangement of putting ground and electric potential on two opposite faces of the material. However, the third arrangement is based on a unique way of electroding, i.e. applying an electric potential (100 V) at the top and bottom faces and providing ground at an orthogonal side, as shown in Figure 11(a). While electric field distribution shows a symmetry about the horizontal central axis of the material, there is a variation of electric field vector from 0° to 90° along the horizontal direction (Figure 11(b)). The maximum amplitude of the

![Figure 11](image1.png)

Figure 11. Pattern III of spatially varying electric field in BST sample, (a) electrode placement, dimensions and boundary conditions of the sample, (b) electric field distribution inside the sample.

![Figure 12](image2.png)

Figure 12. Variations of X and Y displacements, along AA’ and BB’, respectively, for pattern III.
The electric field is at the tip of the ground electrode on the right face of the sample. As the magnitude and direction of the electric field vector show a large variation along X-axis, this pattern should provide a prominent amount of converse flexoelectric coupling in X-direction, meaning a larger magnitude of deformations in the length of the sample, as compared to its height. Thus, the right face of the sample is selected as a target actuation surface for its X-displacement and hence the uniform X-displacement constraint is applied on this face.

At this point, it becomes obvious that pattern III serves the purpose of providing a high amount of converse flexoelectricity, prominently in the X-direction. The linear relationship between electric field gradient and strain yields an actuating effect in the sample. The resulting displacements in X and Y directions are plotted in Figure 12. Y-deformation varies from zero to 1.57 nm, i.e. an elongation in the height of the structure. X-displacement shows a magnitude of displacement of 3.3 nm on the right surface. Thus, pattern III provides an actuation that is higher than the pattern I and pattern II, but requires a special type of electroding arrangement with electric potential at two opposite faces.

Aspect ratio and electrode length ratio significantly affect the output of the two patterns discussed earlier and pattern III is no exception. Hence, we must inspect the effect of these two ratios on maximum horizontal and vertical displacements along the central axes of the structure. Figure 13(a,b) shows the variation of displacements along central axes with varying $R$ and $r$, respectively. Decreasing the aspect ratio increases X-displacement greatly due to the increasing magnitude of the electric field and its gradient. However, as this configuration requires electroding on the thickness of the sample, the decreasing the aspect ratio may pose some difficulties in its practical implementation. The next parameter to be investigated is electrode length ratio, which is the ratio of the length of the ground electrode and the height of the sample, which is plotted in Figure 13(b). A continuous increase is seen in the actuation is

![Figure 13. Variations of maximum X and Y displacements with (a) varying aspect ratio and (b) varying electrode length ratio, for pattern III.](image-url)
observed with increasing electrode length ratio. Horizontal displacement is recorded to have a maximum magnitude of 85 nm, a much higher value in comparison to other arrangements. Similar to pattern II, pattern III also possesses the issue of the coinciding ground electrode and electric potential at higher values of r.

The three patterns proposed in the present study have their own benefits and drawback, depending upon the requirement of the application. For instance, the pattern I may be suitable for thin samples while pattern II and III may be challenging to implement on thin samples due to difficulty in electroding. However, the generation of high electric field gradients without modifying geometry or material composition, through the present method, provides a unique approach for getting improved converse flexoelectricity independent of material and geometry.

4. Conclusions

A novel strategy to induce electric field gradient, and thereby converse flexoelectricity, by varying electric field direction is presented. Three patterns of electric field gradient are obtained by three unique electrical boundary conditions. Pattern I provides an aligned electric field vector, diagonally from maximum electrical potential to ground. This results in a gradient of an electric field to ultimately provide an actuating effect. Pattern II is an extension of electrode placement in the pattern I. The electrical boundary conditions are applied only on horizontal faces in the pattern I, whereas in pattern II similar boundary conditions are applied to vertical faces as well. Although this results in a more uniform alignment of the electric field, it also introduces an elevated electric field gradient in X-direction. Finally, pattern III is introduced which has electric potential at both horizontal faces while grounding on the mid of one of the vertical faces. This arrangement provides an increase in electric field gradient in X-direction. The proposed strategy is established as a solution to the dependence of converse flexoelectricity on the nonregular shape of the structure. Depending on the requirement of the application, any of the three patterns suggested in this study can be employed to generate converse flexoelectricity. This method can be sought as a universal (in terms of shape and material composition of the sample) methodology to have a geometry independent converse flexoelectric effect in dielectric materials. To understand the practicality and feasibility of the proposed approach, parametric studies are conducted for investigating the influences of aspect ratio and electrode length ratio. It is observed that while pattern I can easily be achieved regardless of the dimensions of the sample, patterns II and III pose some challenges when it comes to thin samples.

Disclosure statement

The authors declare that they have no potential competing interest.
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Appendix A

Stiffness matrices,

\[
[K_{qq}] = \int_{\Omega} [B]^T [C] [B] d\Omega,
\]

\[
[K_{q\phi}] = \int_{\Omega} [H_q]^T [\mu]^T [B_\phi] d\Omega,
\]

\[
[K_{\phi q}] = \int_{\Omega} [B_\phi]^T [\mu] [H_q] d\Omega.
\]

Force vectors,

\[
\{F_m\} = \int_{r^1} [R]^T \{t\} ds,
\]

\[
\{F_Q\} = \int_{r^0} [R]^T \{Q\} ds.
\]

Material matrices,

\[
[C] = \frac{E}{(1 + \nu)(1 - 2\nu)} \begin{bmatrix}
1 - \nu & \nu & 0 \\
\nu & 1 - \nu & 0 \\
0 & 0 & \frac{1 - 2\nu}{2}
\end{bmatrix}
\]

\[
[\mu] = \begin{bmatrix}
\mu_{11} & \mu_{12} & 0 & 0 & 0 & \mu_{44} \\
0 & 0 & \mu_{44} & \mu_{12} & \mu_{11} & 0
\end{bmatrix},
\]

\[
[\varepsilon] = \begin{bmatrix}
\varepsilon_{11} & 0 \\
0 & \varepsilon_{22}
\end{bmatrix}
\]