Chapter 2

Literature Review

The literature related to studies on effective properties of piezoelectric composites can be roughly divided into three basic categories: (1) analytical models, i.e., strength of materials (SM), method of cell (MOC), shear lag, Mori-Tanaka approximations, etc.; (2) numerical models based on finite element analysis (FEA); (3) experimental methods, i.e., Berlincourt method, laser heterodyne interferometer, laser-Doppler vibrometry, etc. Based on the above classifications, a literature review of the relevant research papers is being presented in subsequent sections.

2.1. Analytical Models

Most of the analytical solutions concerning the electro-mechanical coupling phenomena in piezoelectric composites involve the derivation of the effective properties, i.e., elastic, piezoelectric, dielectric coefficients of the piezoelectric composites. Classical models, such as the Voigt uniform-strain model and the Reuss uniform-stress model, have been utilized to evaluate effective coefficients on the piezoelectric composites [12]. These analyses are based on the uniformity of stresses and strain produced in both the phases (fiber and matrix). These results were later modified into a more refined model that includes the parallel-serial model [13] and the model developed by [14]. The basic assumptions for refined models remain the same as the classical. Most theories developed to determine effective electroelastic moduli of piezoelectric composites are based on the parallel and series connections of constituent materials depending upon their spatial connectivity characteristics [15]. In earlier attempts, analytical predictions were limited to study of unidirectional continuous fiber reinforced composites where analysis is carried out for only one dimension and proved to be sufficient to yield adequate results [16]. In a more comprehensive approach, [17] developed an analytical model based on the concentric cylinder approach to account for the interactions among the longitudinal continuous fiber at finite concentrations. This model is an extension of concentric cylinder models of elastic and electric behaviour put in combined form to illustrate coupled electroelastic behaviour.

Furukawa *et. al.* [18] derived an expression for the piezoelectric strain coefficient, d_{33} for a particle reinforced composite, i.e., for a discontinuous reinforcement, though their analysis is based upon the assumption of fiber and matrix being isotropic and their behaviour governed by a common elastic, dielectric and piezoelectric constant. This model obtained a reasonable amount of agreement with experimental data, its applicability is severely limited as the results are obtained for only one direction of fiber orientation. Many more attempts were later made to model problems related to discontinuous reinforcement; [19] gave an analytical model based on the generalization of parallel-series model as adopted by [15] (for studying the continuous reinforcement) to consider the effects of discontinuity of the fiber phases in particle reinforced composites.

Wang [20,21] developed a model to obtain expressions for combined electroelastic responses of an ellipsoidal inhomogeneity (fiber) embedded in an infinite matrix. The electroelastic Green's function forms the foundation of the solutions for the combined electroelastic responses in these derivations. Though the solutions for such functions cannot be obtained analytically for anisotropic inclusions, the solutions can be approximated with the use of Fourier transform for certain symmetry groups, e.g., transversely isotropic. Hence, [21] derived explicit expressions for electroelastic coefficients for the case of continuous fiber embedded in a matrix medium. This derivation has certain limitations of being accurate with dilute fiber concentrations only as it ignores the interaction among fibers whose presence ought to be appreciable at finite concentrations. However, the results obtained by [21] for the electroelastic responses is equivalent to the results obtained by [22] through a representation of electroelastic responses in terms of tensors first developed by [23]. Deeg [22] obtained expressions for coupled electroelastic fields for an ellipsoidal inhomogeneity embedded in an infinite piezoelectric matrix through generalization of expressions obtained by Eshelby for elastic responses of the similar problem.

Asaro *et. al.* [24] obtained a solution for eigenstrain in anisotropic elasticity problems which [22] furthered for finding a solution to polynomial eigenstrain and eigen electric field for electroelastic problems. The solutions hence provide expressions for the coupled electroelastic behaviour of the piezoelectric composites. By generalizing the results of [23], Deeg [22] provides an elegant and more widely applicable results as they allow for all possible forms of eigen fields (strain and electric field). A similar treatment of ellipsoidal inclusion problem by [25] provides solutions for the coupled electroelastic fields for the single ellipsoidal inclusion embedded in an infinite matrix. However, there is no attempt to estimate the average fields generated due to the presence of the inclusion.

In the work of [26], the solution given by [22] is utilized to obtain explicit expressions in the form of a set of four electroelastic tensors analogous to the Eshelby's tensor in elasticity for mapping the coupled electroelastic fields. These expressions then are used to derive effective properties of piezoelectric composites in a similar manner for that of [27] analysed such problems in elastic media. The solution obtained thus is generalized through [28] effective medium theory to obtain the estimate for the average electroelastic fields that exist at finite concentrations. The theory proposed by Mori and Tanaka has been effectively used in finding solutions to many inhomogeneity problems linked with uncoupled mechanical [29] and thermal [30] behaviour of composites. However, [26] appears to be the first approach towards the application of solutions to the coupled behaviour of piezocomposites. The solution yields the predictions for effective elastic, piezoelectric and dielectric coefficients of composites reinforced by an ellipsoidal fiber. The choice of ellipsoidal inclusion extends the applicability of the analysis to a wide range of fiber geometries, i.e., spheroid, ribbon shape, thin lamina, penny shape etc as shown in [31]. The analytical results presented are proven to be self-consistent, diagonally symmetric, and exhibiting optimum behaviour in both low as well as high concentration limits.

By systematically treating the integrals derived in [26,32,33] for the piezoelectric inclusion problem, [34] derived the explicit expressions for a cylindrical inclusion (continuous fiber) aligned along axis either parallel or perpendicular to the axis of anisotropy of a transversely isotropic symmetry in a piezoelectric medium. The integral representation of coupled electroelastic tensors is derived for an ellipsoid inclusion in an anisotropic piezoelectric medium by using transformation of the area element that represents the surface of the inclusion. Explicit results for the piezoelectric tensors have been obtained for a spheroid reinforcement in a piezoelectric medium by [35] through deriving Green's functions for transversely isotropic piezoelectric solids. Earlier, [36] used the same Green's function approach for deriving the solution of an ellipsoidal inclusion problem in a transversely isotropic elastic medium.

A significant progress in the field of micromechanical analysis to determine effective properties of piezoelectric composite has been made by Benveniste *et. al.* [37–39]. Benveniste *et. al.* [39] derived the universal relations to correlate the effective coefficients with the constituent properties by extending the uniform-fields concept for the uncoupled mechanical case to piezoelectric phases in binary piezoelectric composites.

By combining the composite cylinder assemblage model with the universal relations derived earlier, the author [40,41] derived some exact expressions for the effective coefficients of the piezoelectric composites that characterize two-phase fibrous piezocomposites.

For a piezoelectric composite having continuous piezoelectric fiber embedded into a matrix, the Mori-Tanaka approach [42], the variational bound technique [43], and the asymptotic homogenization technique [44–46] are most widely adopted approximation techniques used for evaluation of effective properties of piezoelectric composites. Asymptotic homogenization techniques though yield closed-form expressions for prediction of electroelastic properties but the derivation of these expressions involves a very complex mathematical manipulation. The Mori-Tanaka mean field approach is a very efficient technique for prediction of effective properties as the multiparticle interactions of inhomogeneities in a heterogeneous media is captured well in this approach. Different theoretical treatments of the self-consistent schemes which deal with the one -particle problem in heterogeneous medium have been reported in [17,32,47] for estimation of overall properties of piezoelectric composites. Della *et. al.* [48,49] have effectively used the results obtained from the Mori-Tanaka approach to study the performance characteristics of 1-3 piezoelectric composite with and without presence of porosity.

2.2. Numerical Approaches

All analytical and semi-analytical models developed to estimate effective properties of piezoelectric composites eventually reduce to the specific cases of geometric parameters. To avoid the limitations that are tied to specific geometric configurations of analytical models, various numerical models, such as finite element methods, have been proposed over the years. These models add significant contributions to the understanding of overall behaviour of piezocomposites as these models pose no restrictions to geometry, constituent properties, the number and nature (active/passive) of phases and the size of reinforcement. However, the refinement of the results of finite element-based studies are sensitive to the mesh density. So, it becomes a cumbersome task to find the appropriate meshes for analysing problems using FEM. To achieve optimization of mesh size and number a convergence study is carried out for such analysis.

The extension of mean field approach from elastic to electroelastic problems as studied by Wang, Dunn *et. al.*, Benveniste and Chen [21,26,40,50,51] are based on solutions derived from Eshelby type tensors and are found to be quite effective in predicting the entire set of coefficients under arbitrary load. However, the models use average representations of the elastic and electric fields within the constituents of the composite. Unit cell models based on finite element methods are capable of overcoming this restriction. In such models, representative volume elements and boundary conditions are designed in such a way to map specific deformation patterns connected to the applied load cases. These fields are numerically solved with high resolution finite element-based tools, e.g., ANSYS, ABAQUS, COMSOL etc. [52–56]. These approaches allow the prediction of only a few material constants as with symmetric boundary conditions, only normal loads can be applied with consistency. Later, to deal with arbitrary loading scenarios, a different approach was developed by [57,58] named as asymptotic homogenization approach. This approach models the locality in elastic and electric fields and the effective elastic, piezoelectric and dielectric coefficients are measured analytically with hence developed explicit expressions [59].

Poizat *et. al.* [60] determined piezoelectric coefficients d_{11} and d_{33} by applying suitable boundary conditions to the meshed representative volume element (RVE). The unit cell models that predict the entire set of material constants correctly were given by [61,62]. Though in the work of [63,64] Reisner *et. al.* and Bisegna *et. al.* presumably adopted correct boundary conditions; no specific details have been given in the research paper. Berger *et. al.* [65] have used FEM based micromechanical analysis to unidirectional continuous fibre reinforced composites to predict a full set of material constants. The periodic boundary conditions and recurrent loading conditions have been coded with FORTRAN and numerical solution has been performed ANSYS. Few other Agbossou *et. al.*, Xia et. al., Lenglet *et. al.*, Sun *et. al.*, Li, Pastor, Tan *et. al.* and Pettermann *et. al.* [66–73] applied the unit cell approach to capture the local field fluctuations that helps in determining the complete set of material constants that defines overall elastic, piezoelectric and dielectric properties of the piezoelectric materials. These approaches map linear response to any mechanical and electric load, or any combination of both of them. The unit cell approach is an attempt to produce a general procedure for the prediction of effective coefficients of composites with complex geometric reinforcements. For verification of the procedure, different unit cells for square and hexagonal arrangements of the fiber were considered for the study.

The author in [74] advances the theory of unit cell to a more unified micromechanical theory that accounts for the interaction of periodic cells, and this method is quite effective for studying the overall behaviour of composite materials for both, elastic and inelastic constituents. Periodic boundary conditions were applied to the RVE which is valid only for those cases where normal tractions are applied on the boundaries of the unit cell. For a shear loading case, many authors, including Needleman *et. al.* and Tvergaard *et. al.* [75,76], among others, demonstrated that the 'plane-remaining-plane' boundary conditions are over-constrained. In their work [67], Xia *et. al.* further demonstrated that the aforesaid boundary conditions are not only over-constrained but they may also violate the stress/strain periodicity conditions put at the boundaries of the unit cells. Hori *et. al.* [77] presented a universal inequality which suggests that the estimated elastic constants of the composites can vary depending on the applied loading conditions on the boundary of a unit cell. The model also demonstrates that the homogeneous displacement and homogeneous traction boundary conditions will give the limits, i.e., upper and lower bounds, of the effective coefficients.

Hollister *et. al* [78] have given a very good comparison of the homogenization theory and average field theory, concluding that the homogenization theory (uses periodic boundary conditions) yields more accurate results. The authors have also attempted to present a more applicable hybrid theory with the help of correlations that exists between the homogenization theory and average field theory. In [44,46,79,80], Bravo-Castillero *et. al.*, Guinovart-Diaz *et. al.*, Sabina *et. al.* and Rodriguez-Ramos *et. al.* have presented the methods to calculate the effective coefficients of composites with the help of asymptotic homogenization techniques. In a two-part series, each for elastic and piezoelectric analysis has been performed for continuous fiber composite having square and hexagonal arrangement of fiber packing. For elastic problems, the numerical results derived from closed form expressions has been compared with the known bounds of analytical results [81]. For the piezoelectric problem, the numerically computed results have been compared with the experimental data [82] and are found to be in a good agreement.

Kar-Gupta *et. al.* [83–88] have used finite element tools very effectively to study the effects of poling directions (fiber and matrix), complex reinforcement geometries, presence of voids and many such parameters on the effective coefficients, i.e., elastic, piezoelectric and dielectric of the piezocomposites. Kar-Gupta *et. al.* [83] studied the effects of poling characteristics of fiber and matrix on the overall properties of the 1-3 piezoelectric composites. Based on two factors; first the spatial arrangement of the reinforcement and second the relative orientation of poling direction between the two phases; a finite element model has been developed that effectively captures electroelastic response of 1-3 piezocomposites. Similar finite element model has been developed by Kar-Gupta *et. al.* [84] to study the effects of porosity on the electromechanical response of piezoelectric materials. The effects of porosity on the effective properties and electromechanical coupling of the piezoelectric composites have been systematically characterized and results have been demonstrated for the classifications of “longitudinally” or “transversely” porous, depending upon relative orientation of the porosity axis with the principal axis of the poling direction of piezoelectric material.

A three-dimensional finite element has been developed by Kar-Gupta *et. al.* [87] to study the effects of fiber geometries, e.g., laminate, networked, long-fiber, short-fiber, particulate on the electroelastic response of the piezoelectric composites. It has been demonstrated that the geometric connectivity of fiber and matrix phases have significant effects on the performance characteristics of a piezoelectric composite. The derived results also suggest that the grain size modifications of the constituents (i.e., fiber and matrix) can significantly improve the effective properties (e.g., coupling coefficients, charge coefficients, etc.) of a piezocomposite while maintaining appropriate acoustic impedance. Kar-Gupta *et. al.* [87], have presented a comprehensive study of the effects of fiber shape and orientation on the overall properties of the piezoelectric composite. Six different classes of piezoelectric composites constructed with prismatic and non-prismatic fiber shape has been considered for analytical and numerical studies. The effective elastic, piezoelectric and dielectric coefficients have been calculated with an analytical model [89–93] (based on equivalent inclusion method) and a numerical model (based on FEM). The results demonstrate that the effects of the fiber shape on the transverse direction properties is quite significant while the longitudinal direction properties are almost insensitive to any changes in fiber shape.

2.3. Experimental Characterization

As described in literature, experimental characterization of piezoelectric composites can be classified into categories. The first category may be attributed to the measurements of dielectric and piezoelectric constants by using either the direct piezoelectric effect or the resonance method [4]. The second category involves the measurement of surface velocity and displacement at certain frequencies by the application of certain laser probing methods. The author [94] used a Berlincourt piezometer and measured the piezoelectric strain coefficient (d_{33}) value at certain specific locations on the surface of a 1-3 piezocomposite. The average of the measured value at different locations gives the effective piezoelectric constant $\bar{d}_{33}$ of the composite. A static technique based on the direct effect has been used to measure the effective hydrostatic piezoelectric constant $\bar{d}_h$. Similar experimental approach has been adopted by [95] to measure the effective piezoelectric coefficient $\bar{d}_{33}$ for 1-1-3 piezocomposites. Taunaumang *et. al.* [82] have presented an experimental validation to the analytical models by measuring electromechanical properties of the composites. It was found from the study that all electromechanical properties (elastic, piezoelectric and dielectric) agreed with analytical model predictions except the piezoelectric coefficients. The measured values for the piezoelectric coefficients were much lower than what was predicted through analytical models.

Measuring effective piezoelectric strain coefficient based on surface displacement at resonance frequencies was first reported by [96,97] by using a laser heterodyne interferometer. With the help of Laser-Doppler vibrometer, Rittenmyer *et. al.* [98]

performed a direct measurement of the temperature-dependent piezoelectric strain coefficients of composite materials. Zhang *et. al.* [99] reported a direct measurement of the surface displacement profile of a 2-2 piezocomposite at low frequency (200 Hz) using a double-beam laser dilatometer. With the help of a large heterodyne interferometer, the low-frequency performance of a single rod piezocomposite has been investigated [100]. However, to the best knowledge of present authors, characterizing the static deformation in piezocomposites has not been presented in the literature. Chan *et. al.* [101] have extended the analytical model based on the parallel connectivity approach developed by Smith *et. al.* [102] to cover more material parameters and have compared them with the experimental results. The performance parameters, i.e., acoustic impedance, acoustic velocity, electromechanical coupling factors and hydrostatic strain coefficient are predicted through an analytical model and found to be close to the values measured experimentally.

A summary of the analytical and numerical models developed to predict the electromechanical response with respect to different fiber geometries has been given below.

Table 2.1 A summary of the analytical models and numerical models developed to predict effective coefficients of piezocomposites

Classification		**Constituent activity**		**Constituent crystal symmetry**		
	Model	**Fiber**	**Matrix**	**Fiber**	**Matrix**	**Fiber shape**
Analytical	Composite parallel-	Passive	Passive	Transversely isotropic	Isotropic	Long-fiber

	assemblage model					(square, rectangular)
	Composite ellipsoidal assemblage model	Active	Active	Transversely isotropic	Transversely isotropic	Long-fiber (elliptical)
	Models based on dilute, self-consistent, Mori-Tanaka and differential micromechanics theories	Active	Active	Anisotropic	Transversely isotropic	Long-fiber (circular, elliptical) short-fiber (penny shape, spheroidal)
	Asymptotic homogenization model	Active	Active	Transversely isotropic	Transversely isotropic	Long-fiber (circular)
	Double asymptotic homogenization model	Active	Active	Transversely isotropic		Long-fiber (square)

	Analytical models based on Eshelby tensor approach	Active/Passive	Active/Passive	Anisotropic	Transversely isotropic	Long-fiber (elliptical, circular, rod-shaped)
	Isofield based micromechanics model	Active	Active	Orthotropic	Orthotropic	Particulate (ellipsoid, spheroid)
Numerical	Method of cell	Active	Passive	Transversely isotropic	Isotropic	Long-fiber (circular, square)
	Unit cell method	Active	Passive	Transversely isotropic	Transversely isotropic	Long-fiber (circular, square), Short-fiber (ellipsoid)
	Numerical homogenization techniques	Active	Active	Transversely isotropic	Transversely isotropic	Long fiber (circular, elliptical, square)
	Finite element method	Active/Passive	Active/Passive	Anisotropic	Anisotropic	Any possible fiber shape

With the above discussion, it becomes apparent that many micromechanics models are developed over the years (both analytical and numerical) to study the overall behaviour of the piezocomposites. Though many of these models are based on certain assumptions (in each case) which are essential to simplify the complicated mathematical equations. This in turn restricts the applicability of the models to the more complex analysis that involves electromechanical coupled behaviour of piezocomposites. For long-fiber piezo composites, one of the most credible analytical models is based on strength of materials approach, but as Gibson [142] showed for elastic problems, it predicts elastic properties in longitudinal direction (to the fiber) with utmost accuracy but the properties predicted in transverse directions are far from accurate. Later, this analytical model has been applied to electroelastic problems as well for evaluating effective electroelastic coefficients of 1-3 piezocomposites. So, in view of the present authors, the SM model requires a refined approach (similar to Gibson's approach [142] to the elastic problem) while being applied to electroelastic problems related to 1-3 piezocomposites (long-fiber) as well.

The literature review suggests that the electroelastic Green's functions are quite capable of predicting spatial electroelastic coupled responses. But the exact solutions to the coupled electroelastic Green's functions need to be derived in order to study their spatial behaviour (distribution in the space). The solutions to these Green's functions are a precursor to find electro-elastic analogue to elastic Eshelby tensors. With the application of various theories of approximations (dilute scheme, self-consistent scheme, equivalent inclusion method, Mori-Tanaka approx..) the overall electromechanical behaviour of piezocomposites is determined. The exact solutions to the electro-elastic Esheby tensors have been developed in literature but the analysis is quite complex; it requires a simpler approach. First deriving the coupled electro-elastic Green's function solution in exact terms and then applying these equations to derive electroelastic Eshelby tensors would simplify the whole approach.

The study of electro-elastic Eshelby tensors has huge potential when it comes to develop an analytical model that is unified in approach and capable of predicting effective properties of such composites for a variety of length scale and aspect ratio of fibers. The authors feel that this aspect has not been studied in great depth as it should have been. The electroelastic Eshelby tensors combined with approximation techniques (to factor in interactions between multiple inclusions) would certainly help develop an analytical model that is unified, simple and more accurate for predicting the effective electroelastic coefficients of piezocomposites (across the various length scale of fibers). In literature, though various approximation techniques are discussed which are effective in modeling the local fields generated due to the presence of inclusion and/or inhomogeneity and interactions between them. There lacks a comparative study of these techniques that entails which one of these techniques provides the most accurate results.

2.4. Objective of the present thesis

This thesis investigates the effective properties of piezoelectric composites. The work focuses on several topics of practical importance that have not been adequately addressed in the existing literature. These topics include the effects of fiber geometry and packing arrangement on the overall properties of composites; interaction between electric and elastic fields at the local level, especially for short-fiber composites; the optimization of performance parameters by varying interphase properties; and experimental correlation of analytical results derived from various models. The specific objectives are listed below:

- To derive exact solutions for coupled electroelastic Green's functions and study their spatial distribution in and around an inclusion and/or inhomogeneity. It involves solving the integral operator, (Green's function) by parameterizing the geometry of ellipsoidal inclusion over the surface area of a sphere and hence solving the double integral within the prescribed limits.
- Based on these derived exact results for the electroelastic Green's functions, to develop a simplified analytical model to determine electroelastic Eshelby tensors and studying their characteristics across various length scales and geometry of the inclusions.
- Later, with the help of approximation technique, develop a model to account the interaction between inclusions and/or inhomogeneities and combine with the electroelastic Eshelby tensors to derive a unified mathematical formula for evaluating the effective electroelastic coefficients of piezocomposites.
- To study the variation of electroelastic Eshelby tensors with change in fiber geometry and its packing arrangements. Also, to study the effective electroelastic coefficients, (i.e., elastic, piezoelectric, and dielectric) of piezocomposites with respect to change in length scale and geometries of the inclusion and/or inhomogeneities.
- To study the characteristics of short-fiber composites with the derived mathematical formulations from the above analysis. And, to validate the derived analytical model with finite element-based numerical analysis (FEA).
- To present a refined model to strength of materials (SM) for long-fiber composites that is capable of predicting the effective properties measured in the transverse direction (to the fiber) more accurately as the conventional SM model provides

credible results only for properties measured in the longitudinal direction (to the fiber).

- To check the validation of the proposed analytical model, i.e., modified strength of materials (MSM) with another model based on the strain energy conservation and numerical finite element-based analysis (FEA) for long-fiber composites.
- Verification of the predicted results from the proposed model for the various piezocomposites performance parameters, e.g., electromechanical coupling factor, short-circuit stiffness, open-circuit stiffness, acoustic impedance, acoustic velocity, hydrostatic charge coefficients etc. with the experimental results given in the literature.