



*Review*

## **A review on characteristics and recent advances in piezoelectric thermoset composites**

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**Abstract:** Piezoelectric thermoset composites (PTCs) are the class of material having the ability of transformation between mechanical energy and electric energy. In addition to having the advantages of high strength, easier processing, lower temperature, pressure requirement and unlimited storage, PTCs also have high stiffness, high elastic modulus and high strain coefficients. This review presents the advances and approaches used in PTCs and their applications. Various techniques, such as analytical, finite element and experimental methods for analyzing the coupled piezoelectric responses, are also reviewed. This paper also includes current applications of PTCs in strain sensing, vibration control, actuation, energy harvesting, structural health monitoring and biomedical fields. The studies of PTCs and its applications are in the emerging phase, and the review permits to find new notions for interface studies and modelling progresses for PTCs. In addition to that, these reviews pave the way for various research potentials towards the flourishing pertinent application zones of PTCs. Also, this review highlights the relevance of the particular research area and preliminary work under its different approaches, necessitates the need for more researches.

**Keywords:** piezoelectric; thermoset; composite; finite element method; analytical; experimental

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### **1. Introduction**

Composite materials are accessible in many engineering fields like aerospace, automobiles, civil structures, etc., due to their high strength to density and stiffness to density ratios. But nowadays,

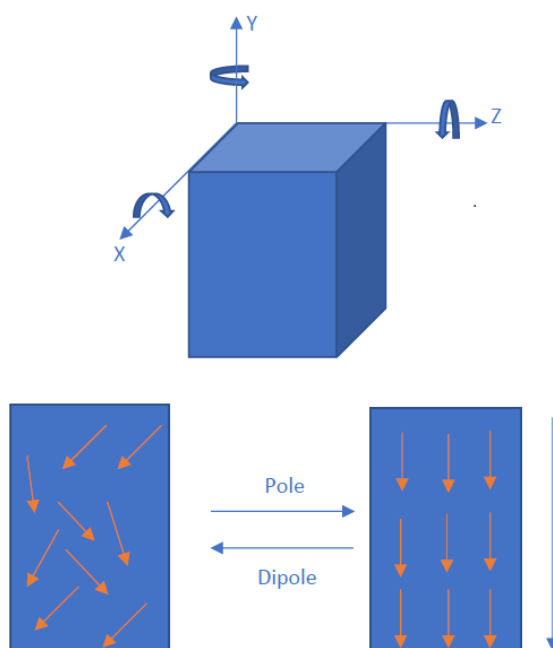
composite materials being the primary load-carrying member of any structure is expected to have additional functionality along with its primary services. These other functions enable the composite structure to sense and to provide actuation to the system under consideration and control the same. The conventional composite materials can refine their properties and be smart by joining in hand with adaptive, and smart materials. The most common smart materials are piezoelectric and electrostrictive materials, shape memory alloys, magnetostrictive materials, electro and magnetorheological fluids, fibre optics etc. When these smart materials are bonded or embedded in composite materials, the performance and capabilities of the conventional composites are improved and acquire some additional functionality to its primary (usually structural) purpose. Smart materials can respond to changes in stress, strain, displacement, velocity, acceleration, electrical, thermal or other mechanical change of a structure through changes in their properties in a controlled manner to maintain desirable and satisfactory performance.

Among these smart materials, piezoelectric ceramics as sensors and actuators exhibit unique and superior characteristics over the others which are suitable for applications like noise and vibration, shape and position control, non-destructive testing, energy harvesting and health monitoring systems. The fields of applications of piezoelectric thermoset composites (PTCs) ranges from, aerospace industry to micro and nano-electromechanical sensors and actuators. So, piezoelectric ceramics, act both as sensors and actuators in PTCs. This paper also reviews piezoelectric ceramics and its applications in biomedical, aerospace, automotive, sport and machine tool industries, aviation, civil structures etc., due to their lightweight, self-monitoring and self-controlling capability.

## 2. Piezoelectric elements and characteristics

Piezoelectric ceramics and piezo polymers are active materials which possess the ability to alter the geometric to material properties under the application of various external stimuli, thereby acquiring an inherent capacity to transduce energy [1,2]. In short, in piezoelectric elements generates piezoelectric and the inverse piezoelectric effects. That is, the generation of electric charge upon piezoelectric material is deformation and development material deformation upon application of electric charge. The dipole re-orientation of piezoelectric material by an external electric field is shown in Figure 1.

Though the most common piezoelectric ceramics are Lead zirconate titanate (PZT), Lead magnesium niobate (PMN), Lead meta niobate (LMN) and Lead titanate (LT), PZT's have been the most widely employed. The most commonly used piezo-polymer is Polyvinylidene fluoride (PVDF) even though other piezo-polymers like Polyvinylidene trifluoroethylene P(VDFTrFe), polyvinylidene fluoride tetrafluoroethylene and their copolymers exist [3]. Various piezoelectric materials and their strain capabilities are specified in Table 1.



**Figure 1.** Dipole re-orientation by an external electric field in the piezoelectric material.

**Table 1.** Piezoelectric materials and their strain capabilities.

Piezoelectric material	Density (g/cm <sup>3</sup> )	Piezoelectric (strain) constant $d_{33}$ (m/v)	Electro-mechanical coupling coefficient $k_{33}$	Refs.
PZT	7.80	$4.50 \times 10^{-10}$	0.66	[4,5]
PVDF	1.78	$4.00 \times 10^{-10}$	0.12	[6]
PMN-PT	7.70	$3.57 \times 10^{-10}$	0.94	[6,7]
LMN	6.30	$0.77 \times 10^{-10}$	0.42	[8]

### 2.1. Lead zirconate titanate (PZT)

Combining the chemical compound titanate with lead and zirconium under very high temperature result in the formation of PZT. Superior material characteristics like high dielectric constant, high coupling coefficient and high density along with fine grain structure and noise-free response make PZT highly employable inflow or level sensors, ultrasonic non-destructive testing, generation of high-voltage energy in ultrasonic cleaners, sonar devices, etc. In addition to this, higher mechanical quality and more economical operation increase its demand among other piezoelectric materials [4–7].

The PZT samples(a  $\text{PbTiO}_3$  and  $\text{PbZrO}_3$  mixture) exhibits high values of electro-mechanical coupling factor and energy conversion effectiveness coefficient depending on the composition [8]. Electro-mechanical coupling factor,  $k_p$ , is an indicator of the energy conversion effectiveness and piezoelectric charge constant.  $d$ , is the mechanical strain experienced by a piezoelectric material per unit of the electric field applied. The high values of  $k_p$  and  $d$  in PZTs make it highly acceptable than the other piezoelectric ceramics. This PZT system embedded over polymer composites were studied by several investigators [9–14].

## 2.2. Lead magnesium niobate-lead titanate (PMN-PT)

PMN-PTs which finds its application as actuators demonstrate large electrostriction strains [15]. These materials exhibit outstanding piezoelectric properties (e.g.,  $d_{33} = 357 \times 10^{-12}$  m/v,  $k_{33} = 0.94$ ) and it has other specific advantages such as negligible hysteresis, lower creep and lack of high voltage that considerably outdoes PZT ceramics [16–17] but exhibits inferior piezoelectric performance. PMN-PTs finds its application as actuators in the area of biomedical for endoscopy [18] and as hearing aid [19]. PMN-PTs are challenging to manufacture and lead titanate ceramics in bulk form undergo a phase transformation, and this limits its application [20].

## 2.3. Lead meta niobate (LMN)

In pure form, LMNs exhibits the most substantial piezoelectric anisotropy [21] but are of limited efficacy practically because of the difficulty in manufacturing. This piezoelectric phase experiences severe cracking while production [22]. Numerous studies had done to fix this issue by introducing various additions but had limited success, leading to a sharp decrease in anisotropy [23]. Due to this drawback, practical applications of LMNs are limited.

## 2.4. Polyvinylidene fluoride PVDF

PVDF is the most common piezoelectric polymers used as sensors and actuators. PVDF polymer is soft and flexible and therefore can be attached easily on to the curved surfaces. Further, PVDF is chemically inert, robust, creep resistant, and has excellent stability when exposed to sunlight [24–26]. Also, it has a low density along with low dielectric permittivity resulting in a very high voltage coefficient [27,28].

But low  $d_{33}$  and  $d_h$  values and little dielectric constant inherent in PVDF have limited its use. In addition to this, the poling the PVDF film is a touch difficult due to the requirement of the high electric field. Despite their high dielectric breakdown values, the low piezoelectric voltage constant makes them inapt for energy harvesting applications [27].

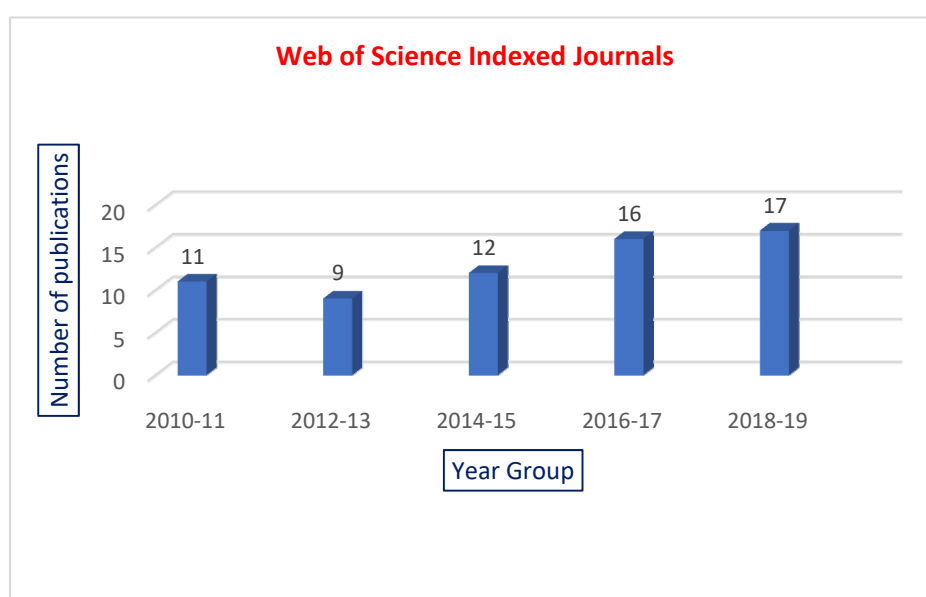
## 3. Necessity and trends in piezoelectric composites

Embedding piezoelectric materials in composites depend on the required properties of smart composite structures. Several factors such as type and dimensions of piezoelectric material, its position, its compatibility with the composites, etc. play a vital role in choosing and embedding the right kind of piezoelectric materials for composites.

Even though the excellent properties of PZT have widens its area of applications, its unfortunate mechanical strength act as a limiting factor for their life cycle and performance. Also, the brittleness, inflexibility and high densities of the piezoelectric ceramics lead to considerable acoustic resistance which necessitates the need for matching layers [28]. Piezoceramic materials also add additional mass and stiffness to the host structure, especially when working with flexible and lightweight materials. In such a status quo, fibre-reinforced polymer composites can be used as a matching layer for piezoelectric materials which improves the load-bearing capacity of the piezoceramics along with retaining all the qualities of conventional ceramics (electrical, mechanical, chemical). Coupling

piezoceramics and polymer composites mitigate the weight and brittleness issues of piezoceramics. Besides, this enhances the overall mechanical properties and strength of the composite [29].

Therefore, interest in polymer-ceramic [30–34] composites have emerged as an area of interest [35,36]. These composites can be polarized under the influence of an external electric field [37]. Piezoelectric ceramic/polymer composites possessing numerous association outlines had been explored in several studies during the past several years [38,39]. The relationship between the number of Web of Science-indexed publications under the area of piezoelectric ceramics over the last few year groups, is shown in Figure 2. The surge in the number of articles portrays the relevance of the field and the developments occurring in this area. In the present review, the focus is mainly oriented towards the work done in polymer composites due to its better strength and stiffness properties. The strength and stiffness properties within the plane of polymer composites can be controlled more precisely.



**Figure 2.** Details of Web of Science-indexed piezoelectric composite articles.

#### 4. Approaches in smart composites

The main approaches in smart composites can be classified as analytical, numerical and experimental. For the proper characterization of composite materials and their interfaces, the use of multiple analytical techniques is required. Analysis of composite materials can be quite complex, and composite material analysis laboratories need correct analysis tools to characterize and resolve many of the problems of composite materials.

An extensive review of different analysis approaches, namely, analytical approach, finite element approach and experimental approach on piezoelectric thermoset composites are depicted in this paper.

#### 4.1. Analytical approach

Smart composites can be investigated using mathematical models when largescale experimental studies are expensive and challenging to conduct. Geometrically imperfect piezo-magnetic nanobeams were analyzed in [40,41] to investigate the thermal post-buckling behaviour. In that work, the nanobeams were considered as functions of piezoelectric phase percentage and reported that the increase in phase percentage had increased the applied voltage sensitivity meanwhile increase in nonlocal parameters resulted in lowering the post-buckling temperatures. Ganesh et al. [42] had analyzed a delaminated composite plate with active fibre composite under hygrothermal environment. They have also considered the effect of moisture and temperature and observed that the natural frequencies were reduced due to delamination. A complete dynamic analysis and significant coefficients were extracted from reinforced piezo-magneto-thermo-elastic plates by Hadjiloizi et al. [43] for a set of unit cell problem. According to them, the significant coefficients were not a constant but a function of time. A mathematical model of a piezoelectric sensor was developed by Asif et al. [44], to study the debonding effect and verified that the developed model could recover the presence and extent of partial debonding between the composite laminate and the piezo sensor.

The static, free vibration, dynamic control and transient characteristics of piezoelectric laminated composite plates were analyzed in [45–47]. And a novel solution for finding twisting deformation and optimal shape control of smart laminated composites plates was developed by Soheil et al. [48]. From the study, it was observed that the laminate stiffness could distress the twisting bending coupling developed by the inclined piezoelectric actuators. The major works using analytical approaches for piezoelectric polymer composite is shown in Table 2.

**Table 2.** Summary of major analytical approaches for piezoelectric polymer composites.

Composite material	Piezoelectric material	Approach	Properties	Refs.
Laminae	PZT-5H	Extensional Hamilton's principle and improved layer theory	Degrading performance of the partially debonded sensor	[15]
Piezo-magneto nanobeam		Hamiltons principle	Thermal post-buckling	[41]
Graphite epoxy Wafer reinforced magnetoelectric plate magnetoelectric shell with honeycomb filler	A.F.C. layer	Potential energy approach Maxwell's equation	Parametric study Significant coefficients and dependent field variable	[42] [43]
Elastic layer	Piezoelectric + Piezomagnetic layer	Maxwell's equation	Mechanical and electrical properties	[44]
Graphite epoxy	PFRC actuator	Navier's method and Principle of virtual work	Transient characteristics	[45]
Graphite epoxy	PZT + PVDF	Classical variational formulation	Static and dynamic vibration control	[46]
GFRP		Kirchoff's hypothesis	Twisting deformation	[47]
Graphite epoxy	CFRP	Higher-order shear deformation kinematics	Flexural behaviour	[48]

#### 4.2. Finite element approach

Among the various approaches available, the finite element approach is the useful preliminary tool to analyze the effect of multiple parameters on PTCs. Several studies enumerating the static,

dynamic, linear, non-linear parametric analysis using the finite element approach were cited by many researchers [15,45,48–55]. While the integrity limit and failure behaviour of piezoelectric sensor embedded fibreglass composite was studied by Lampani et al. [56], the degree of polarisation and piezoelectric characteristics of PVDF integrated kevlar-carbon fibre composite was investigated by Michael et al. [57]. The optimal shape control and twisting deformations of PTCs were investigated by Soheil et al. [48] and concluded that the usage of inclined piezoelectric actuators could suspend the pure twisting deformation. Mehrdad et al. [58] had modelled an active composite strut and an active composite panel to find out the optimum voltage for vibration suppression was modelled by Dutta et al. [59]. The primary studies with finite element methods for piezoelectric polymer composite are summarised in Table 3.

**Table 3.** Summary of central finite element works in piezoelectric polymer composites.

Composite material	Piezoelectric material	Approach	Properties	Refs.
Laminae	PZT-5H	FEM (Extended Hamilton's principle and improved layer theory)	Dynamic characteristics	[15]
Graphite epoxy	PZT + PVDF	FEM	Static and dynamic vibration control	[46]
GFRP	KYNAP	FEM-Abaqus (Kirchoff's law)	Twisting deformation and optimal shape control	[47]
Graphite epoxy and CFRP	Trefnol-D + PZT-4	FEM-Ansys (Higher-order shear deformation kinematics)	Flexural behaviour	[48]
CFRC	PZT	XFEM-Abaqus (Galerkin's method)	Tensile and in-plane shear properties	[49]
Aluminium boron fibre	Piezoelectric + Piezomagnetic layer	FEM-Ansys (Classical laminate theory and Viscoplastic theory)	Mechanical properties and non-linear responses	[50]
Graphite epoxy	PZT	FEM (Variational principle)	Material nonlinearity	[51]
Graphite epoxy and glass epoxy	PZT	FEM-Ansys	Vibration	[52]
Graphite epoxy	Trefnol-D	F.E.M. (Third order shear deformation theory)	Non-linear static behaviour	[53]
Graphite epoxy	PZT-4	FEM (Virtual work principle)	Static parameters	[54]
Graphite epoxy	PZT-5A	FEM (Maxwell's equation)	Static- nonlinear	[55]
Fibreglass	PZT	FEM (Strength-based approach)	Damage	[56]
Kevlar carbon fibre	PVDF	FEM-Ansys (Dunn and Taya micromechanical approach)	Degree of polarization and mechanical and piezoelectric characteristics	[56]
Composite strut and composite panel	PZT-5A	FEM	Optimum voltage	[58]

### 4.3. Experimental approach

Various experimental studies in the field of piezoelectric composites are included in this section for a better understanding of the influence of multiple static and dynamic parameters of composites and to validate various finite element and analytical models. The tensile, in-plane shear and bending properties of PZT piezoelectric embedded carbon fibre reinforced composite was studied by Swati et al. [49] for analyzing progressive damage. The mechanisms of damages are explored by Lampani et al. [56] with the help of the four-point bending test of fibreglass composite embedded with PZT piezo element. Tao et al. [60] had done an investigation to improve the load-bearing capacity of

PZT/PVDF composites and concluded that the addition of aramid fibre could cause a substantial improvement in the same. The relevant research works using experimental approaches for piezoelectric polymer composite is included in Table 4.

**Table 4.** Summary of major experimental works in piezoelectric polymer composites.

Composite material	Piezoelectric material	Properties	Refs.
CFRC	PZT	Mechanical properties (Electronic Universal Testing Machine)	[49]
Fibreglass	PZT	Mechanical and electrical capacity (4-point bending test setup)	[56]
Aramid	PZT + PVDF	Mechanical and electrical properties	[60]

## 5. Applications of piezoelectric composites

Piezoelectric embedded composites become acceptable only if its structural integrity is promised. The reinforcing fibres should be disturbed minimum, and mechanical properties of the composite should not be reduced [60]. Piezoelectric composites find its applications in various fields of engineering.

Ariel et al. [61] had used flexible solar cells into the compliant wings of a Robotic bird (Flapping Wing Ariel Vehicles, FWAVs). FWAVs are comprised of a carbon fibre -mylar composite. Various wing designs have been suggested by integrating a diverse number of solar cells in different positions on Robotic bird. Integration of solar cell has increased the stiffness of the wings, and the deformation produced during flapping generates aerodynamic forces for the flight. Also, the use of solar cells increases the payload capacity by electrical energy harvesting. Thus the addition of solar cells makes the wings multifunctional by allowing it to produce electrical power, senses the changes in wing deformation and to harvest solar energy during flight. This advanced technology is used in aerospace and its applications have been explained in [62].

Another central area where PTCs find its application is in the advancement of morphing aircraft wings and its review was done by Thill et al. [63]. The study reveals that the composites and polymers with structural flexibility and elastomers with a low cross-link density were considered for morphing since they can undergo sizeable elastic deformation without permanent changes. Integrated composites with SMA wires and Fibre Bragg Grating (FBG) were also considered and were used as self-actuating structures. Even though the study discloses the difficulty to combine the properties like flexibility and stiffness to one design, it highlights the aerodynamic performance and operational benefits of morphing technology and the possibilities of PTCs in morphing technology.

A piezoelectric composite actuator was designed and validated by Mudupu et al. [64] for a smart projectile fin. The piezo-fibres, embedded within the epoxy matrix and coated with skin, were used to design a fuzzy logic controller for the fin. The study presented the robustness of design to overcome various disturbances and subsonic wind velocities. Another application of integrating electronic communication antenna into a composite structure was detailed in [65–68]. The design and fabrication of a microchip antenna over a three dimensional orthogonal woven composite structure [69], its impact testing [70] and damage analysis of the composite [71] had been recently developed in the field of aerospace.

The application of polymer composites with graphene-silver hybrid nanoparticles in the biomedical field was presented by Kumar et al. [72]. The uniform distribution of the nanoparticles



helps better exfoliation and dispersion of the nanofiller in the polymer composite matrix. It also improves the storage modulus and enhances electrical conductivity.

The applications of luminescent ions- in the composite matrix and other advanced composite materials were reviewed by Bai et al. [73]. Luminescent ion coupled composites have additional optical and electrical properties, and hence they find its application in sustainable energy devices for solar light harvesting. Also, their better optical properties have been exploited for diagnosis, nano-composite biosensors and medical treatment in the biomedical field.

Biranche et al. [74] had reviewed the critical characteristics and fabrication routines of piezoelectric materials, including piezoelectric ceramics, piezoelectric polymers and piezoelectric composites from the perspective of bone tissue engineering. The relatively high piezoelectric properties of ceramics combined with bioactive fillers enhances the potential of ceramic matrix composites as bone tissue engineering material. But the major limitation of this is the absence of mechanical simulation control.

Vaidya et al. [75] had developed a woven E-glass with vertical piles model which enhances the impact resistance and vibration damping. A 3D space available configuration of sandwich material is used in form filling, which has increased the strength of the laminate and the impact load-carrying capacity. Experimental analysis for the active vibration control of E-glass/epoxy laminated composites using piezoelectric ceramic patches was done by Song et al. [76] an analytical study of the same combination was done by Bohua et al. [77]. Thierry et al. [78] had conducted a vibration reduction study to increase the lifespan and avoid the fluttering in the composite fan blades of a turbojet engine using piezoelectric devices.

Structural health monitoring of cement-based composites with embedded piezoelectric ceramic had been studied by Biqin et al. [79]. A novel cement-based piezoelectric sensor to the in-situ stress-time history monitoring of a reinforced concrete frame has been developed, and the results revealed the feasibility and applicability of the same in the concrete structures. Vibration analysis on the impact response of multi-layered cement-based piezoelectric composite was done by Taotao et al. [80].

The application of damage detection in smart panels composites had been illustrated by Phong et al. [81]. In the study, a non-linear vibro-acoustic wave modulation technique was used for the damage analysis of carbon/epoxy smart composite panels. The research includes the stationary statistical characteristics of vibroacoustic responses. A theoretical model of carbon nanotube reinforced piezoelectric cylindrical composite shell has been considered by Hossein et al. [82–89]. The effects of transverse shear and rotary inertia have been included in the study. The wave propagation characteristics are investigated considering the impact of various piezoelectric coupling factor, different polarization and different orientation of the nanotubes. The investigation of the effects of nanotube agglomeration on wave propagation and vibrational characteristics in hygrothermal were also included in the study. The proposed models find its application in dynamic stability analysis as well as in structural health monitoring as non-destructive testing. Table 5 includes significant works in various application fields of the piezoelectric polymer composite.

**Table 5.** Summary of applications of piezoelectric polymer composites.

Material	Application	Characteristics	Refs.
Carbon fibre with Mylar	Solar energy harvesting	Stiffness, payload capacity	[61]
Composites with SMA wires and FBG	Morphing aircraft wings	Structural flexibility, low cross-link density	[63]
Epoxy matrix with piezo-fibre	Smart projectile fin		[64]
Polymer composite with graphene-silver hybrid nanoparticles	Biomedical field	Storage modulus, electrical conductivity	[72]
Composite matrix with luminescent ions	Solar light-harvesting, biomedical field	Optical and electrical properties	[73]
Ceramic-composite with bioactive fillers	Bone tissue engineering	Piezoelectric property	[74]
E-glass/epoxy composite with piezoelectric ceramic patches	Vibration control	Impact resistance	[76–78]
Carbon epoxy smart composite	Damage detection	Vibro-acoustic properties	[81]
Piezoelectric cylindrical composite with carbon nanotube	Structural health monitoring, dynamic stability	Wave propagation characteristics	[82–89]

## 6. Directions of future research

The review reveals that significantly less number of investigations have been enduring under experimental methods due to the requirement of highly expensive equipment and difficulty in manufacturing. In these scenarios, analytical and numerical methods found most promising in the analysis of PTCs. An extensive scope of study exists in the investigation of various types of piezoelectric materials, their coupling effects on thermoset composites, its failure modes etc. The various fields of application of PTCs are also wide open for further studies. PTCs finds its application in the emerging fields of energy harvesting, structural health monitoring, biomedical fields etc. and the research potential in these areas are in the nascent stage. So, this review paves the way for new studies in the promising regions of PTCs and its applications. Also, the increase in the number of studies in the specified area as from Figure 2 reveals the relevance of the topic.

## 7. Conclusion

In this paper, an overview of various piezoelectric materials and multiple approaches in PTCs are presented. PZTs are found to be the most promising piezoceramic though studies are going on to improve the electrical and mechanical characteristics of other piezoelectric materials. When used with a matching layer, the functionalities of both the piezoelectric element and the attaching layer can be improved, and thermoset composites are found to be best suited for this purpose. The applications of PTCs are not limited to the fields of aerospace, structural health monitoring etc. but to vibration control, energy harvesting, biomedical application and many more. A comprehensive review of the related journals covering approaches and applications in PTC is done here. The study reveals the need for extensive development in the area of piezoelectric thermoset composites as the range of applications is broad.

## Conflict of interests

All authors declare no conflicts of interest in this paper.

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