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# Magnetostrictive polymer composites: Recent advances in materials, structures and properties

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# ABSTRACT

Magnetostrictive polymer composites (MPCs) are a class of materials having the ability to simultaneously change dimensions, elastic and/or electromagnetic properties under the presence of a magnetic field. Their advantages over bulk magnetostrictive metals are high resistivity, extended frequency response, low weight, ease of formability and improved mechanical properties. In this review, advances in MPCs and their applications since the year 2000 are presented. A wide range of reinforcements and morphologies used to generate magnetostrictive response in polymers are considered, including carbonyl iron, nickel and rare-earth metal based reinforcements. A critical analysis of the various polymeric systems from stiff thermosets to soft elastomers is provided, focusing on how the material selection influences the magnetorheological and magnetoelectric properties. Multiscale approaches, such as continuum micromechanics based theories and multi-physics finite element approaches, for modeling the coupled magneto-elastic responses are also reviewed. Recognizing their unique properties, recent applications of MPCs in electric current and stress sensing, vibration damping, actuation, health monitoring and biomedical fields are also presented. The survey allows us to shed light on new directions for fundamental research, interface studies and modeling improvements for advancing the application of MPCs.

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Nomenclature					
d <sub>33</sub>	piezomagnetic strain coefficient				
$E_3^{\tilde{H}}$	Young's modulus at constant magnetic field				
ψ	strain sensitivity $(d\lambda/dH)_{\sigma}$				
$\dot{\lambda}_{\parallel}$	axial magnetostriction (in magnetic field direction)				
$\lambda_{\perp}^{''}$	transverse magnetostriction (perpendicular to magnetic field direction)				
ω	volume magnetostriction ( $\approx \lambda_{\parallel} + 2\lambda_{\perp}$ )				
AC	alternating current				
DC	direct current				
CFO	CoFe <sub>2</sub> O <sub>4</sub>				
CPVC	critical particle volume concentration				
CV	coefficient of variation				
$\alpha$ and $M_0$	Langevin function fitting parameters				
DMF	N,N-dimethylformamide				
EMD	easy magnetic direction				
HFGMC	high fidelity generalized method of cells				
MDE	magnetodielectric effect				
ME	magnetoelectric effect				
MPC	magnetostrictive polymer composite				
MR	magnetorheological effect				
PMMA	poly methyl methacrylate				
PDMS	polydimethylsiloxane				
PVDF	polyvinylidene fluoride				
PVDF-TrFE	polyvinylidene fluoride-co-trifluoroethylene				
PZT	lead-zirconate titanate				
ROM	rule of mixtures				
1-D	Tertenol-D				
VOI%	volume fraction (in percentage)				
wc	tungsten carbide				
wt%	weight fraction (in percentage)				

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# 1. Introduction

The development of magnetostrictive materials is promising for futuristic electromechanical devices with higher energy density, faster response and superior precision. When using giant magnetostrictive materials such as Terfenol-D (T-D) which is a magnetostrictive alloy of terbium, dysprosium and iron, it is possible to realize high strain and energy density for low frequency and high power applications. However, the monolithic T-D materials are not easy to be molded into desired shapes for producing devices; they are brittle and can only withstand small strains to failure. Consequently, research quickly began to progress to investigate how giant magnetostrictive materials can be combined with polymer matrices to create Magnetostrictive Polymer Composites (MPCs). The benefits of using MPCs are not only in reducing the rising costs associated with dwindling supplies of rare earth metals, but also in overcoming the problems in operating monolithic-magnetostrictive devices at frequencies higher than 1 kHz. Combining magnetostrictive materials within polymer matrices allows for improvements in the ability to mold to the desired structural shapes [1–4].

When magnetostrictive particles are surrounded by a polymer with a high resistance, the dynamic response is improved. The insulation provided by the typical epoxy binder influences the distribution of eddy current by reducing the impact especially at high frequencies [5]. Using the polymer in the composites can increase the response up to 100 kHz which is much higher than the sub kHz observed in the monolithic magnetostrictive materials [5]. MPCs also offer the flexibility in tuning their responses by aligning the giant magnetostrictive materials in the desired direction. In addition, MPCs have higher stress and strain to failure compared with their monolithic counterparts, an important issue when the devices are operating at resonant frequencies and experiencing high strains. Unique magnetostrictive responses in composites can also be accomplished by using magnetorheological elastomers.

In this review, we examine the progress in MPCs by surveying the literature post the year 2000 to understand the current state of science with constituent materials development, processing, structure, modeling and applications of MPCs. Previous reviews related to this topic have primarily investigated applications of monolithic magnetostrictive materials [6–8], magnetoelectric composite materials [9–13] and magnetorheological elastomers [14,15]. This review for the first time presents a study of MPC considering the various combinations of magnetostrictive materials and the polymers for magnetostrictive, magnetoelectric and magnetodielectric applications. We examine the reinforcement effects from a wide array of materials including rare earth materials which are inherently capable of changing volume or the composites which use carbonyl-iron that can be activated to change the volume of the composite under applied magnetic fields. The MPC are examined independently in thermosets, thermoplastics or elastomers considering the effects of the polymers on the overall response under coupled electromagnetic, mechanical and thermal fields.

#### 2. Physical and chemical characteristics of magnetostrictive constituents

Ferromagnetic materials display an interesting behavior called magnetostriction which causes a change in volume and thus strain when subject to a magnetic field. The response can reach up to 1 percent strain depending on the chemistry of the magnetostrictive alloy and the content of rare earth materials included. The fields of energy harvesting and sensing has been quick to seize on the opportunities of wireless application provided by giant magnetostrictive materials. These fields have been dominated with the giant materials despite interesting properties from other materials such as cobalt ferrites or nickel. Application of magnetostrictive materials and especially composite based ones is a complex optimization problem involving the need to account for stress, temperature and magnetic fields. The typical composition and some relevant properties of most common magnetostrictive (inter)metallic alloys typically used in MPC can be seen in Table 1.

# 2.1. High magnetostrictive Terfenol-D alloys

Terfenol-D is an alloy of rare earth materials and iron that is synonomus with high magnetostriction materials (Fig. 1). These materials belong to a class of alloys that can reach strains values between 1000 and 2000 microstrain (i.e. 0.1–0.2%) corresponding to a magnetic field in the 50–200 kA/m range under different temperatures [22,23]. The variability reported in the literature is in the order of five percent for the saturation strain. [24]. The strain experienced can be influenced by the temperature of the material especially for the case of the saturation strain [25]. The strain in the magnetostric-tive materials can also be anisotropic with significant property improvements resulting if there is a preload applied. The direction of the preload has a significant impact on the magnetostrictive response [26].

## Table 1

List and characteristics of active materials typically used in M	PC
--	----

Material	Typical composition	Saturation strain, $\mu$ s	Density (kg/m <sup>3</sup> )	Magnetic permeability	References
T-D alloy	Tb <sub>0.3</sub> Dy <sub>0.7</sub> Fe <sub>2</sub>	1200-1700	9200	2-5	[6,16]
Galfenol alloy	Fe <sub>81.6</sub> Ga <sub>18.4</sub>	200-250	7800	75-100	[16,17]
Cobalt ferrites	FeCo	100-150	2400-3100	2	[18,19]
Carbonyl iron	Fe	-	7860	21.5	[20,21]



Fig. 1. The wide and sharp magnetostriction responses attributed to the anisotropy [29].

The magnetostrictive response can be traced to the spin-spin and spin-orbit interactions with the intermetallic phase having the ability to undergo changes in the crystalline structure [27]. An enhanced magnetostriction is observed with T-D with higher proportions of Tb to Dy which results in an increase of magnetocrystalline anisotropy [28]. A small negative magnetostriction is observed after overcoming the coercive field. Domain rotation and shape anisotropy can explain a "W" shape magnetostriction when an axial load is present [29] at small magnetic fields. Of interest in many practical applications are the "V" and "U" shapes observed for parallel and transverse magnetic fields as shown in Fig. 1. Volume magnetostriction has also been measured and it presents a negative value that is a function of the preload. Its magnitude has been reported from 200 ppm at zero stress to 1000 ppm at compression stress values of 50 MPa [30]. The strain derivative is an important parameter for device design and can be influenced by the use of a bias field and be improved with preload [31].

Magnetostrictive rare earth alloys such as T-D have the highest saturation strain levels but they also use expensive rare earth metals. Transition metal alloys offer economical alternative materials that can still be used to fabricate MPCs (for e.g. the Fe-Ga single crystal has a magnetostriction over 200 ppm [32]). The large shape anisotropy of spherical particles caused by the high saturation magnetization of transition metals makes it difficult to achieve magnetic softness. Other studies have reported use of other rare-earth elements as substitution for Dy and Tb, like Pr can impact the solidification process [33]. The fabrication of T-D and other magnetostrictive alloys frequently results in anisotropic properties as they require fabrication of single crystals or polycrystalline materials. A crystal oriented at [111] will yield significantly greater magnetostriction than a polycrystal without any preferential orientation. Polycrystalline MPC manufactured with needle shaped [112] yields significantly more magnetostriction shows that shape anisotropy can be more important in some cases than magnetocrystalline anisotropy [34].

# 2.2. Galfenol alloys

The Fe–Ga (Galfenol) alloys can show high amounts of magnetostriction (not as high as T-D), but need a lower field for saturation. The Galfenol alloys have a low saturation field and a high permeability along with good mechanical properties (Fig. 2). These behaviors have moved the alloys of Fe-Ga and Fe-Ga-Al to be considered for various MPC that can promise good mechanical properties and lower costs than T-D based ones. Although having a lower saturation magnetostriction than T-D, it possesses better mechanical properties such as higher strength and ductility. The room temperature saturation magnetostriction exceed 300 ppm and are only weakly temperature dependent [32]. Binary Fe-Ga alloys with a maximum tensile stress of 200 MPa and failure strain of 0.18% can be increased by about 3 and 5 times by the addition of TaC carbide reinforcements [35]. Similarly NbC doping has been reported to increase the tensile strength to 620 MPa and the strain at break to 0.9% [36]. Rare earth additions can increase the properties, but that is also likely to increase the price which is one of the motivations for moving away from T-D into using Galfenol. Dy added into a Fe<sub>83</sub>Ga<sub>17</sub> alloy enhanced the magnetostriction to 300 ppm at 400 (kA/m) magnetic field [37].

#### 2.3. Cobalt ferrites

Cobalt ferrites while not showing as much magnetostriction have been proposed as alternatives to T-D and other magnetostrictive materials. The typical composition of cobalt ferrites are reported in Table 1. These materials show almost linear magnetostrictive behavior until 225 ppm [38]. The processing of cobalt ferrite has shown that the largest magnetostriction occurs at low temperatures applied for extended dwells [39]. A major drawback with these materials is that they are limited to be operated above sixty degrees because of the hysteresis at lower temperatures. However research shows that some alloying with iron may increase the Curie temperature [38]. Fe<sub>1-x</sub> Co<sub>x</sub> alloys with greater than 50% Co showed the largest



Fig. 2. The magneto-mechanical response of a single crystal of Galfenol (100) [32].

magnetostriction of 108 ppm [40]. In thin films, Hunter et al. reported effective magnetostriction greater than 260 ppm under a magnetic field of  $\sim$ 10 mT [41].

# 2.4. Carbonyl-iron

The typical properties of carbonyl iron are shown in Table 1. These materials are typically associated with the magnetorheological and magneto-active MPCs with an elastomeric matrix [42–47]. In fact, magnetostriction can also be accomplished when using non-magnetostrictive iron powders when low modulus matrices such as elastomers are used. When these materials are used, interaction between particle/particle and particle/matrix interaction become critical. These materials can result in large magnetostriction values (>9%) much larger than those of other MPCs [48]. Active powders function to introduce magnetic effects and also to reinforce the polymeric matrix. In general, larger particles of 50–60 µm [43], improve the behavior; however the modulus can be maximized when smaller particles, in the 3–10 µm range, are used [45,49,50].

# 2.5. Other magnetostrictive materials

Materials that do not comprise rare earth alloys or use them to a lower amount have also been researched due to the cost benefits possible and unique properties. Introduction of Pd to iron alloys has shown magnetostriction of approximately 650 ppm due to microstructural changes that can be introduced by controlling the solidification process [51]. The magnetostric-tive susceptibility of a Fe–Pd alloy film was higher than a film produced from a T-D alloy film when magnetic field is between 25 and 250 Oe (1 Oe  $\approx$  79.58 A/m) thus offering possibility of low noise operation and better control at low magnetic fields [52]. While most MPC reported in the literature possessed positive magnetostriction, other materials had intrinsic negative magnetostriction [53].

# 2.6. Reduction of particle size for composite applications

There have been several studies on understanding the performance of MPCs, particularly on the effect of particle distributions. The packing density has a crucial effect on the magnetostriction saturation and response in MPC response [1]. MPCs containing particles with a size ranging from 100 to 300  $\mu$ m produced larger magnetostrictive strains, whereas those with smaller particle sizes (<100  $\mu$ m) yielded higher compressive strengths but reduced magnetostriction [1,5]. This peculiar behaviour could be ascribed to the higher filler/matrix interfacial area developed when the particle size is reduced. Fe<sub>3</sub>O<sub>4</sub> nanoparticles synthesized by a solvothermal procedure and an oxidative hydrolysis method resulting with particles ranging from 9 to 50 nm average size with magnetostriction ranging from 26 to 167  $\mu$ c [54,55]. The size and volume fraction of the reinforcements in MPC can impact response at high frequencies [56]. Alignment effects of the particles in MPCs showed that the orientation of the magnetostrictive fillers exhibit larger magnetostrictive relative to randomly oriented particles [57].

Additional work on the orientation effect has been performed using crystallographically aligned  $\langle 1 \ 1 \ 2 \rangle$  magnetostrictive particle composites [58–60]. Using flake type particles with tungsten carbide (WC) and aligning them can help in achieving good properties under a relative low magnetic field [61]. Fig. 3 shows that flake or flat shaped particles show a better response than rounded shapes [62]. Therefore, a positive effect on the magnetostrictive response can be detected when the active phase is either oriented or shaped with a favorable morphology (i.e. high shape factor).

Various methods have been reported for producing magnetostrictive particles from FeGa, suction castings followed by ball milling [63], gas atomization [64], spark erosion [65], forging [40], and mechanical alloying. Mechanical alloying of Fe and Ga powders milled for time periods of up to 6 h converts the elemental constituents to an alloy dominated by the chemically disordered A2 phase yielding a maximum of magnetostriction of  $\sim 41 \,\mu\epsilon$  observed in Fe<sub>81</sub>Ga<sub>19</sub> composites (similar to polycrystalline FeGa) [66]. Milling over 6 h results in a large drop in magnetization with increasing milling time, consistent with the appearance of the secondary L1<sub>2</sub> FeGa phase. Pulverization of Galfenol (Fe<sub>80</sub>Ga<sub>18.5</sub>Cu<sub>1.5</sub>) into coarse pieces of several millimeters and then ball milling for four hours have been reported to obtain flake type Galfenol powders. The addition of copper in the alloy facilitates the fabrication of flake-type particles (1–3  $\mu$ m thickness) because the copper atoms have high tendency of segregation at the grain boundaries [67]. Oleic acid has been proposed for surfactant assisted high-energy ball milling to produce Tb0.3 Dy0.7 Fe1.92 nanoflakes with a thickness of 50–400 nm [68]. The alloy subjected to high-energy ball milling with 15% weight of oleic acid developed into nanoflakes with an increased coercivity of 3.8 kOe (1 Oe  $\approx$  79.58 A/m) and its magnetization decreased. This increase of magnetic coercivity may be directly associated with the reduction of grain size (15–21 nm).

Nanostructured MPC (piezo polymer shell with magnetostrictive core) are predicted to produce superior performance because the small thickness of the piezoelectric polymer shell relative to the core. The magnetostrictive stress will be absorbed by the whole volume of the piezoelectric phase (not just the surface), thus generating a larger relative response. Better contact between the interfaces will ensure proper transformation of deformation between the phases. Nanostructured fibers have been developed showing a linear change in the piezoelectric response of CFO/PVDF nanofibers [69]. This composite was produced with an average fiber diameter of  $\sim$ 325 nm and showed a change in the piezoelectric response of these electroactive nanofibers when a magnetic field is applied [69]. Multiferroic nanocomposites based on three different ferrite-nanoparticles,  $Zn_{0.2}Mn_{0.8}Fe_2O_4$  (ZMFO),  $CoFe_2O_4$  (CFO) and  $Fe_3O_4$ (FO), dispersed in a PVDF-TrFE matrix also showed a high dependence on the filler content on the ME effect [70]. Mainly the FO and ZMFO showed no hysteresis behavior. Magnetoelectric CFO/PVDF microspheres prepared by an electrospray process have also been proposed for use in ME applications. Concentrations of CFO nanoparticles in the microspheres reach values up to 0–27 wt% [55].

Nanosheets with a magnetostriction of 507 ppm have been synthesized by a co-precipitation method using a modified gas-slugs microfluidic system. The microfluidic approach enables to grow anisotropic FeCo based nanostructures in a reproducible and continuous fashion under a time scale of 1 min [71]. Nanosheet fillers in different alignment states (random, transversal, and longitudinal) of  $\delta$ -FeO(OH) have been introduced into piezoelectric P(VDF-TrFE) polymer matrix composites [72]. The authors observed a new ME effect based on the rotation of the  $\delta$ -FeO(OH) nanosheets inside the polymer. Even if the strains obtained were small (0.51 ppm) the approach is promising for application in microelectronic devices where detection of magnetic fields is necessary.



Fig. 3. Magnetostriction vs magnetic field for composites fabricated with various powders at a fixed compaction pressure of 0.5 GPa in Fe-Co composites [62].

## 3. Magnetostrictive polymer composites

In a MPC, the magnetostrictive particles will undergo magnetostriction in a polymer matrix in response to a magnetic field. The particles will apply forces on the polymer matrix resulting in an overall deformation of the composite. Equilibrium is achieved by balancing the stresses generated in the particles and the polymer resulting in total deformation of the composite. The microstructure is the primary driver of the overall response of magnetostrictive polymer composites [58,60] and a large part of this section will consider the different effects of microstructure on the overall response. The interaction between the geometry and stress has been found to significantly impact the magnetostrictive properties. Both the processing and the mechanical properties of materials (magnetostrictive particles and polymeric components) will govern the response of the MPC.

For strength and low-temperature formability thermosetting matrices are preferred. On the other hand, thermoplastic resins are suitable for a variety of processing techniques such as injection molding, blow molding, extrusion and thermoforming. Therefore, thermoplastic polymers have also been used as matrices in MPCs and will be discussed. In order to maximize the deformation of the MPC, elastomers can also be used to provide a large stretch. In the following sections, we discuss the trajectory of research on the above topics and other emerging ones. Since the measurements of applied field vary widely among various research groups, it is believed by the authors that including the slope of the strain vs. magnetic field curve in the review would lead some confusion in the comparison of material performance. Hence, the saturation magnetostriction is used to present and track the performance of composite materials. Table 2 summarizes the current MPC capabilities reported across the literature.

#### 3.1. Microstructured materials

In general, the higher concentration of magnetostrictive material so is the magneto-elastic response. However, there have been several studies showing that this may not always be the case [1]. When one set of particle sizes are used (i.e. monodisperse), the 250–300  $\mu$ m MPC produces the highest strain response, but due to inefficient packing this MPC also shows the lowest increase in strain for increasing preloads [1]. The larger particle performance is attributed to the minimum surface area of the larger particles and the minimization of demagnetization. Using a bimodal particle distribution it is possible to manufacture a composite where demagnetizing effects are minimized and packing density is maximized [1].

The magnetostrictive properties of T-D/epoxy tend to increase with increasing particle size, but a high compressive strength is obtained at low particle sizes [5]. The particle size and volume fraction effects on dynamic properties show that eddy current reductions increase the operating frequency of MPC [56]. Improved magneto-mechanical properties are the result of alignment of crystals in the MPC [58–60]. Typically, there are two types of MPC composites, namely the 0–3 MPC when dispersing magnetostrictive particles in a polymer matrix and 1–3 MPC that involve aligning the particles in the matrix of the composite. The EMD of Dy-rich alloys prefers to lie along  $\langle 1 0 0 \rangle$  axis, whereas that of the Tb-rich alloys favors to lie along  $\langle 1 1 1 \rangle$ , which can be ascribed to their different anisotropies of DyFe2 and TbFe2, whose EMD lies along  $\langle 1 0 0 \rangle$  and  $\langle 1 1 1 \rangle$ , respectively [73,74]. Magnetically aligned particles in chain structures can be best classified as 1–3 composites [75]. Microscopic analysis of MPC specimens with polyamine cured (low stiffness) and anhydride cured (higher stiffness) epoxy matrices reveals different types of defects in the microstructure [76]. The damage visible in the polyamine-cured composite is in form of extensive cracking both inside and near the interface. The anhydride-cured composite also exhibits particle internal and near interfacial cracking but to a lower extent. The internal cracking or interface degradation may lead to reduced cyclic magnetostriction.

Epoxy-based MPC composites with Tb<sub>0.32</sub>Dy 0.68 (Fe0.8Co0.2)2 [74] were reported to have higher longitudinal magnetostriction than previously reported T-D/epoxy composites [77,78]. The magnetization curves for the 1–3 composites were above the curve for the 0–3 type, indicating the easily saturation for magnetization, which can be attributed to EMD  $(1 \ 1 \ 1)$  of the textured particles lying along the direction of the magnetic field. In general, aligned magnetostrictive composites perform better than non-aligned ones. The critical value when the orientation field becomes insignificant has been reported to be about 80 kA/m [79], although this may be a function of the volume fraction and polymer characteristics. The particles rotating towards the field direction during the manufacturing process produce a 1-3 composite compared to the 0-3 composites produced without the aligning field. Composites have also been made with T-D slit into long fibrils having a diameter of 1000 µm and length of about 3000 µm and also showing excellent saturation magnetostriction values [60]. While the majority of thermosetting work has been performed on epoxy, there was some work done using phenol binders. Shin et al. [80] used flake-type powders of Fe<sub>36</sub> Co<sub>62</sub> Ge<sub>2</sub> with an average size of 73 µm and with a thickness of 3-5 µm prepared through a conventional arc-melting and ball-milling processes. These particles were mixed at 10 wt% with a phenol binder and compacted under constant pressure of 0.67 GPa. Saturation strains of 89–105 ppm are reported in this type of composites [61,80]. MCP using a phenol matrix and Co ferrite powders below 50 µm were manufactured using a press approach [81]. The MCP had reduced properties compared to monolithic materials but had an interesting property of a wider stress sensing range. The stress sensing capability was also found to be dependent on temperature as a large increase of relative permeability was detected when the temperature was increased. The effect was less pronounced when a compressive stress was applied.

#### Table 2

Magnetostictive properties of MPC materials reported in the literature.

Composite/polymer	Particle size	Particle content	Saturation Magneto- striction ( $\times 10^{-6}$ )	Comment	Ref.
Nickel (hollow or solid sphere)/ vinyl ester	<25 μm	24 vol%	-24 to -28		[53]
Galfenol flakes/epoxy	75–225 μm	40 wt%	10-14		[67]
Sm <sub>0.88</sub> Nd <sub>0.12</sub> Fe <sub>1.93</sub>	10–180 µm	50-60 vol%	-0.62	Alignment at 180 kA/m	[127,128]
CoO-Fe <sub>2</sub> O <sub>3</sub> /phenol	<45 μm	3.5-3.8 wt%	38		[81]
Fe-Ga/epoxy	20–25 μm	48 wt%	53.5	18.9% Ga	[65]
Galfenol particles/epoxy	<25 μm	69.1 vol%	60	Compacted at 271 MPa and aligned at 2 T	[64]
Fe <sub>81</sub> Ga <sub>19</sub> /epoxy	<25 μm	69.1 vol%	60	-	[64]
Silicon steel in/silicon rubber	0.15–0.2 mm	40-80 vol%	100	Random	[107]
Carbonyl iron/silicon rubber	5 µm	27 vol%	134	Alignment at 40 kA/m	[45]
Carbonyl iron/silicon	5 µm	30 wt%	9% strain	Dependent on shape, cylinder with aspect ratio of 0.33	[48]
Fe <sub>80</sub> Ga <sub>20</sub> /epoxy	50–100 μm	80 vol%	360	Compressed at 120 MPa	[63]
Tb <sub>x</sub> Dy <sub>0.9-x</sub> dNd <sub>0.1</sub> (Fe <sub>0.8</sub> Co <sub>0.2</sub> ) <sub>1.93</sub> / epoxy	<150 μm	20-27 vol%	390 (at ~3-6 kOe)	Alignment at 8–10 kOe. x = 0.3–0.4	[129,130]
Tb <sub>0.32</sub> Dy0.68(Fe <sub>0.8</sub> Co <sub>0.2</sub> ) <sub>2</sub> /epoxy	60–120 μm	27 vol%	550 (at ~3 kOe)	Alignment at 10 kOe	[74]
T-D/epoxy	5–300 µm	70 vol%	720 (at 9 MPa)	Using axial force and alignment	[93]
$Tb_{0.3}Dy_{0.7}Fe_{1.7}/epoxy$	Fiber type (20 mm long)	-	761	At 80 kA/m and 10 MPa	[131]
T-D/epoxy	-	20 vol%	800 (at 7.6 kOe)	Static alignment at 6 kOe	[98]
T-D/polyurethane	0–300 μm	50 wt%	813	Oriented	[83]
T-D/epoxy	<300 μm	20 wt%	900 at 0 MPa	Alignment at 2 kOe	[78]
T-D/epoxy	30–500 μm	50 vol%	~1000	Treated with titanate coupling agent and aligned	[100]
Tb <sub>0.3</sub> Dy <sub>0.7</sub> Fe <sub>2</sub> /epoxy	210 µm average size	77 vol%	~1005	1000 MPa compaction. d33 $\sim$ 4.08 $\times$ 10 $^{-9}$ m/A	[95,132]
T-D/epoxy	-	20 vol%	1050 (at 7.6 kOe)	Dynamic alignment at 4 Hz at 6kOe	[98]
T-D/epoxy	45 mm long fibrils	50 vol%	1270	112 composite	[133]
T-D/polyurethane	212–300 μm	50 wt%	1390	Alignment is at 0.5 T	[125]
T-D/vinyl ester	<1000 μm	49 vol%	1475	Aligned particles	[34,59]
Tb <sub>0.3</sub> Dy <sub>0.7</sub> Fe <sub>1.90</sub> /epoxy	200–300 μm	40 vol%	679 at 0 MPa to 1358	Alignment at 8 kOe	[77]
	•		at 17 MPa preload	-	
T-D/epoxy	0–180 μm	97% by wt	850-1000 (at ~5 kOe)	Alignment at 2 T, warm heating and 154 MPa	[94]
T-D/epoxy	<1000 µm fibrils (3:1 ratio)	35-49 vol%	1500-1600		[58,60]
T-D/epoxy	Bimodal 5–50 μm and 250–300 μm	20 wt%	990 at 0 MPa to 1100 at 10 MPa preload	Alignment at 2 kOe	[1]

Polyurethane as a binder has also been investigated for a range of studies in MCP [82–88] including foam based MPC [42]. Thermoplastic polyurethane have many desirable properties, including elasticity, transparency, and resistance to chemicals. Kiseleva et al. [87] used Fe–Ga alloy particles in a polyurethane composite with particles synthesized using a mechanochemical approach [89] to obtain polycrystalline particles of less than <2  $\mu$ m. Using a 25 wt% fraction the authors were able to obtain a saturation magnetostriction of approximately of 1  $\times$  10<sup>-4</sup> at fields above 3–4 kOe (1 Oe  $\approx$  79.58 A/m) for oriented particles with a significant difference between aligned and non-aligned particles. In foam applications, lightweight MPC with low-density and improved compressive properties can be realized [42]. Some research has been reported using carbon black or carbon fibre in polypropylene with a melt-mixing and injection molding process [90]. The authors reported a saturation magnetostriction of 1163 ppm at 800 kA/m that is found to be dependent on time. We have not been able to find other confirming studies.

Kubicka et al. [91] produced 0–20, 20–100 and 100–300  $\mu$ m particles by means of a sieving procedure. The reader is referred to this publication for an excellent assessment of grinding and sieving methods. Fine sieving was performed using an airjet screening machine with a 20  $\mu$ m sieve size. A three-roll mill is used to reduce the particles even further in the range of 15 and 10  $\mu$ m and for particles below 10  $\mu$ m, the bead mill is found to achieve the desired results. The reference provides further details about the settings used for the milling process. Laser diffraction has been used to quantify the distribution of the particles after particle size reduction methods. The HELOS BR system for example can analyze cumulative distributions of particles with sizes between 100 nm and 875  $\mu$ m [91]. The higher the concentration and the smaller the particle size, the larger will be the number of chains. In addition, the lower the distance between the chains produced better homogeneity of the composite [91]. The particle sizes are also found to affect the magnetic field by

concentrating the field responsible for the formation of the chain structures. Smaller particle sizes reduced the distances between the chains (Fig. 4).

Under a certain magnetic field the modulus decreases as the magnetic field increases due to transverse domain motion (e.g. 100 kA/m [58]). When this phenomenon is exhausted, the modulus increases as the magnetic field increases beyond a point due to the pinning of parallel domains by the applied field. High concentration of T-D (greater than 70% volume fraction) in an MPC was investigated with a force-assisted method before curing to displace large amounts of epoxy from the specimen [93,94]. With this method, a magnetostriction of 720  $\mu$ ε was reported. In another approach, a warm compaction was applied at elevated temperature (130 °C) under a 154 MPa pressure and a 2 T oriented magnetic field along the length dimension of the sample, and it resulted in strains up to 1000  $\mu$ ε [94] for 97% weight fractions. The saturation and piezo magnetic response was maximum when the approximately 200  $\mu$ m the particles were aligned yielding a strain of ~1000 ppm and 4 nm/A [95].

#### 3.2. Effects of preload and temperature

When cured under alignment field of 8000 Oe and significantly high compressive prestress, directionally solidified oriented crystals of T-D magnetostrictive alloys exhibit enhanced magnetostriction [96]. Similarly, the optimized magnetostriction in composite material is increased nearly by 100% from  $679 \times 10^{-6}$  at 0 MPa to  $1358 \times 10^{-6}$  at 17 MPa [77]. The magnetic properties can also be dependent on the film/substrate due to residual or thermal stresses. With the increase of the tensile prestress, the magnetization decreases and reaches a saturation level in high magnetic field [97]. Du et al. [98] used a dynamic orientation process by oscillating/spinning the T-D/Epoxy particulate composite along a cylindrical axis during the pre-cure stage. The T-D particles were found to aggregate in several planes to form parallel layers. As a consequence, the magnetostriction of the dynamically oriented composites with laminated structures was increased.

MPC tested from -40 °C to 40 °C show similar reduction in modulus at constant field (18%) for a 50% volume fraction with increasing temperature (similar to monolithic materials) [2]. Testing under elevated temperature shows that  $E_3^H$  is temperature dependent in the range of ±40 °C from zero due to the polymer. Results for the constant load test under thermal loading indicate a peak in the magnetostriction between 0 and 10 °C at all levels of preload for the 50% volume fraction composite. These results also show that the maximum magnetostrictive strain decreases significantly at temperatures below 0 °C due to easy axis switching (with peak between 0 and 10 °C). The magnetostriction of MPC correlates well with monolithic results where a peak in strain occurs at 0°C [96].



**Fig. 4.** Effect of particle sizes on the magnetic flux line density. The first column has no reinforcement (control). The middle column shows impact of large particle sizes less than 300 µm and the third column shows the impact of smaller particles less than 15 µm [92].

# 3.3. Effect of particle treatments

The magnetostrictive particles can be obtained in a variety of sizes and then even further processed. According to the information available in the open scientific literature, to achieve a good adhesion seems not to be an issue. The effects of oxidation while understood on the conductivity of nanoparticles [99] appear to be less clear on the magnetostrictive properties of the MPC.

Magnetostrictive composites prepared with (titanate coupling agent) treated particles present higher magnetostrictive properties, especially at low field levels [100]. The bond strength may explain the increased transfer of the magnetostrictive effect into the polymer phase. Pretreatment of the T-D particles with a silane coupling agent increases the percolation threshold or the concentration of T-D particles. However, it also creates an inactive soft interfacial layer that can impact magnetoelectric properties at higher concentrations of T-D [101].

# 3.4. Giant magnetostriction using elastomers

Thermoplastic polyurethane elastomers are multi-phase segmented polymers that display excellent mechanical and elastic properties, good hardness, as well as high abrasion and chemical resistance. They can also be coupled with nonmagnetostrictive iron particles to produce large (giant) magnetostrictive deformations larger than 9% [48]. They are block copolymers whose chains are composed of alternating "rigid" urethane which have a glass transition temperature, Tg, above room temperature, and 'soft' segments with a lower Tg [102]. Elastomers with a very large strain capability are very attractive for applications in which high degrees of stretching and magnetic response are needed. Filled with certain amount of magnetic particles (even if not magnetostrictive by themselves), an elastomer can be made to display magnetostrictive behaviour. These composites are also referred to as magneto-active or magneto-rheological (MR) elastomers or sometimes referred to as magneto-active elastomer (MAEs). In these composites, a dispersion of particles is subjected to a magnetic field during curing to align the particles in the direction of the magnetic field. Once the polymer is cured that structure is maintained. The effect of this orientation results in a typical modulus dependence on magnetic field. Magnetostriction in these composites can be explained by the presence of small layers of elastomer between the particles [103]. The interaction of the magnetized particles through the elastomer matrix is the source for changes in dimensions and magnetostriction in these materials. The behavior of MR elastomers under compressive, tension and shear have been previously reported in a number of studies [104–106].

During curing of MR elastomers, a magnetic field of 0.1–0.2 T is applied to align the particles. The magnetostriction of carbonyl iron particles dispersed in a silicone rubber matrix) shows that magnetostriction increased with larger volume fraction of particles oriented along an appropriate direction [45]. The magnetostriction increased with rising magnetic field after initially low rate of increase, i.e. sensitivity, for low field. In addition, after repeated cycling the saturation strain decayed and some remnant magnetostriction existed after switching off the magnetic field in each cycle.

The initially low sensitivity region can be reduced with increasing particle density. In composites, the "burst strain" occurs when the alignment changes from random to ordered with the magnetic fields [107]. This is then followed by saturation. On the other hand, removal of the magnetic fields results in a large hysteresis that may be explained by interface effects between the particles and the polymer as no relaxation was observed after the removal of the magnetic fields.

Improved mechanical properties are realized by silane treatments of particles [108], silicone oil additives [46], or addition of carbon black [44,109]. The effect on MR is not conclusive, as it does not improve the MR effect in all cases, especially when the particles are uniformly distributed. There is a belief that in some cases there is a plasticizing effect on the MR elastomers which makes the matrix softer and thus helps to improve the MR effect [108]. Most rubbers contain fillers, particulate solids, which are embedded into the elastic matrix fillers that correspondingly increase the area between loading/unloading curves contributing to the hysteresis losses [43]. The normal force is highly dependent on the magnetic field more than other properties [110]. Rheological properties are dependent on the distribution of the magnetic particles in the composite. The size and connection of particles is directly proportional to the high strains and stiffness values [43].

Magnetostrictive gel specimens were also made of silicone gel with 80% weight fraction of 3.8 µm embedded carbonyl iron particles. At a strain of 30%, the compressed pure silicon experienced a stress of 13.8 kPa, while for the particle filled gel the stress rises to 75.2 kPa [111]. Through alignment of embedded carbonyl iron particles (volume fraction of 27%) relative to pure silicone, the tension, and compression properties were more than doubled or tripled in some cases [47]. Exceptionally tough and notch insensitive magnetic hydrogel was reported by dispersing alginate-coated Fe<sub>3</sub>O<sub>4</sub> nanoparticles into the interpenetrating polymer networks of alginate and polyacrylamide, with hybrid physical and chemical crosslinks [112]. Strip of un-deformed magnetic were stretched to 8–11.0 times the initial length without rupturing including with the presence of notches.

Soft magnetic particles were also assembled in a polydimethylsiloxane (PDMS) matrix to fabricate new MR elastomers with uniform lattice and BCC structures using 400  $\mu$ m iron spheres placed through a layer by layer approach using a laser etched metal mold with variable spacing illustrating how specific microstructures can be obtained [113]. The Payne effect (drop in viscoelastic shear modulus under cyclic loading conditions with small strain amplitudes) was not found to be significant in many studies [114–116]. However, one study suggested that coating the particles with PMMA results in a larger storage modulus and a smaller loss factor. The materials prepared with the coating particles have a lower MR effect, they have weak Payne effect and small steady loss factor which is attributed to the increased bond strength and less relative

motion between the particles and the polymer [117]. Structured materials also show less damping than unstructured materials [118].

Stiffening and magnetostriction both occur when a magneto-active elastomer is placed in a magnetic field [119]. The coupling between this magnetic field and mechanical response in a the composite is determined by the magnetization energy in the material and its strain response [49]. Regarding the MR effect (or stiffening with magnetic field), a higher curing field leads to a material with a stronger stiffening effect [120]. Carbon black is also found to even out the inner stresses in the elastomeric rubber and lets more molecular chains effectively carry the load [109]. Pseudo-plasticity and high fold increases in the MR effect can be achieved by using softer elastomers which can facilitate the particle reorganization under applied fields [121]. The possibility of coupled sensing incorporating magnetostriction and resistivity properties is potentially possible using nickel particles embedded in an elastomer whose electrical resistivity changes by many orders of magnitude under small compressive strain making it a very sensitive pressure sensor [103].

Lokander showed that the MR effect and other properties (e.g. strength) may be limited by a "Critical Particle Volume Concentration" (CPVC) which can be calculated from the "apparent density" of the powder to that of the solid material [122,123]. Benefits of magnetic alignment may not be realized after this concentration (approximately 30% for iron), after which all the particles are touching each others and are surrounded by a continuous polymer matrix. If the air of voids is replaced totally by polymer, the result is a composite filled with a critical amount of particles. At low volume fractions there is more polymer between the particles whereas the higher volume fraction results in situations where there are more gaps because the polymer cannot fit all the gaps. The CPVC approach may run into issues when there are particle agglomerations as in the case of carbonyl iron and using smaller particle sizes.

The higher concentration requires a longer time for hardening due to a delay in the cross-linking [124]. The composite was also found to have a less desirable distribution at the higher loading levels. The magnetic particles can prevent curing and therefore the elastic modulus will also be reduced [124]. The highest MR effect has been found for samples with carbonyl-iron particles  $6-9 \mu m$  in diameter [50] with the highest yield strength at  $30^{\circ}$  with respect to the magnetic field. Inclusion of a small amount (<1%) of 10- $\mu m$  particles in a composite largely consisting of 40  $\mu m$  particles (~30%) was found to enhance the MR response possibly due to the beneficial effect of smaller particles sitting in interstitial positions.

In a study of various T-D particle sizes in a polyurethane elastomer it emerged that the largest particle size with narrowest distribution provided the highest magnetostriction effect [125]. The oxidative stability of natural rubber was found to decrease dramatically (from chemiluminescence and oven ageing) at high volume fractions due to the impact of iron ions on the decomposition of the rubber considering the large amounts of oxygen that maybe present on the particles [126].

Table 3 reports the current MPC reported during dynamic testing. The effects of eddy currents in composites are greatly reduced and allow extension of the operating frequency range 200 kHz [56,134]. The *magnetomechanical coupling coefficient*,  $k_{33}$  is one of the fundamental parameters characterizing the dynamic response of magnetostrictive materials and it quantifies the electromagnetic energy converted into mechanical energy. The magnetomechanical coupling coefficient,  $k_{33}$  can be calculated from the elastic moduli measured at constant magnetic field,  $E_3^H$  and the constant magnetic flux density,  $E_3^B$ , which can also be determined from the resonant  $f_r$  and anti-resonant  $f_a$  frequencies by the following equation:

$$k_{33} = \sqrt{1 - \frac{E_3^H}{E_3^B}} = \sqrt{1 - \left(\frac{f_r}{f_a}\right)^2}$$

The piezomagnetic coefficient,  $d_{33}$  is also used to characterize the dynamic properties and is linked to the measured dynamic strain,  $S_3$  using:

$$d_{33} = \frac{S_3}{H_3}$$

The low values of k in MPC arises from the low polymer modulus. Use of a higher stiffness polymer results in higher coupling values and also low operating strains [135]. Incidentally, the particle sizes do not impact the dynamic coupling constants and coefficients, whereas the dynamic relative permeability is primarily proportional to the volume fraction of the magnetostrictive particles [56]. The coefficient,  $d_{33}$  of the MPC has been found to vary significantly as a function of temperature with a peak typically occurring when the modulus is lowest [2]. The addition of WC to the MPC augments mechanical properties and increases the resonant frequency [136]. Experiments have revealed a strong dependence of composite

 Table 3

 Dynamic properties of various MPC reported in the literature.

Polymer	Magnetostrictive material	Particle content (vol%)	Critical H <sub>bias</sub> (kA/m)	max d <sub>33</sub> (nm/A)	Source
Vinyl ester	112- needle shaped Tb <sub>0.30</sub> Dy <sub>0.70</sub> Fe <sub>1.92</sub>	49	30	5.5-14	[3,58]
Vinyl ester	Ball milled Tb <sub>0.30</sub> Dy <sub>0.70</sub> Fe <sub>1.92</sub>	51	30	3.3	[3]
Epoxy	Sm <sub>0.88</sub> Nd <sub>0.12</sub> Fe <sub>1.93</sub>	51	180	-0.49 to -2	[127,139]
Epoxy	(Tb <sub>0.3</sub> Dy <sub>0.7</sub> ) <sub>0.75</sub> Pr <sub>0.25</sub> Fe <sub>1.55</sub>	50	140	3	[138]
Epoxy	T-D	61	65	1.8	[56]

dynamic properties relative to the availability and motion of non-180° domain states within the magnetostrictive active region of the MPC. Increasing the field above a critical value enables more domain wall motions, which overcome stress-induced anisotropy and decrease the bias field required to maximize the properties [137].

Lv et al. [138] studied a pseudo-1–3 MPC with epoxy and 0.5 volume fraction aligned magnetostrictive (Tb, Fe and Dy) particles with addition of Praseodymium (Pr) in the 10–300 µm range. The composites show no frequency dispersion (between 25 Hz and 70 kHz) except for the resonance range. The dynamic relative permeability demonstrates a decreasing trend with increasing Pr due to the weakening of magnetization saturation with increasing Pr content. The modulus is also affected by the increased Pr compliance.

The piezomagnetic coefficient is a function of the volume fraction and the preload. The higher the volume fraction the higher is the coefficient. A higher compressive load results in a reduction of the coefficient since the domains will be mainly transversally oriented for the composite compared to the case of lower loading in which both parallel and transverse domains are present [58]. Fig. 5 shows how temperature can influence this coefficient which is a function of the stiffness relation to temperature. The damage after dynamic loading can be in the form of particle or interface cracking [76]. The internal cracking or interface degradation will lead to reduced cyclic magnetostriction.

The largest  $d_{33}$  and  $k_{33}$  values observed at a critical bias field are a result of maximizing non-180° domain-wall motion occurring from 0 to this critical bias field value. The composite shows a reduction of modulus to this point. A magnetic domain is a region within a magnetic material in which the magnetization is in a uniform direction. The magnetostrictive material may have multiple domains oriented in different directions separated by domain walls. The domain wall represents a transition between different magnetic moments and usually undergoes an angular displacement of 90° or 180°. The non-180° domain states are related to residual compressive stresses in the composites from processing. Above this value of this critical bias field, the non 180° domain wall motion is constrained and the composite stiffens again [134]. Various values of the critical bias field are reported in the literature, e.g. 30 kA/m for  $\langle 1 1 2 \rangle$  aligned needle type T-D composites [3], or 140 kA/ m in Pr containing particles [138]. Using [1 1 2]-aligned needle shaped particles, resulted in 67% increase in  $d_{33}$  compared to irregular shaped particles showing the importance of shape anisotropy. Sm<sub>0.88</sub>Nd<sub>0.12</sub>Fe<sub>1.93</sub> has been used for negative magnetostriction dynamic studies [127]. The dynamic relative permeability exhibits a flat frequency response with no observable dispersion at all bias field levels, except for the fundamental shape resonance range of 40–50 kHz.

# 3.5. Magnetoelectric and magnetodielectric MPCs

Since PVDF has strong piezoelectricity with the piezoelectric coefficient much higher than any polymer, it has found interest for use in ME MPCs. PVDF is a thermoplastic fluoropolymer. It is easily processable into various monolithic shapes or foams because of its relatively low melting point of around 177 °C. Moreover, it is attractive for biomedical applications because of its biocompatibility [140,141]. The magnetostrictive particles can be dispersed in a polymer solution and then films of different thicknesses can be obtained after solvent evaporation [142]. In the case of ME composites, the ME effect or the induced electric polarization under an applied magnetic field is quantified by the magnetoelectric voltage coefficient:

$$\alpha_{ME} = \frac{V}{H_{AC}d}$$

where *V* is the recorded voltage,  $H_{AC}$  is the amplitude of the sinusoidal magnetic field and *d* is the thickness of the specimen. The induced voltage can be measured by a lock-in amplifier operated in differential mode whereas the applied magnetic field can be measured by a Hall probe or other magnetic measuring techniques. The use of PVDF matrix has been reported in two and three phase materials with magnetostrictive and piezoelectric particles for enhanced ME effects [143–149]. Zeng et al. studied the effect of the magnetic field on the coupling coefficient in PVDF based ME composites [145].  $\alpha_{ME}$  was found to be impacted by the induced magnetic field during the hot pressing process. PVDF has also been used as a binder for laminated T-D composites with PZT [150]. The researchers laminated the T-D/ PVDF and PZT/PVDF particulate composite layers and then molded the composite together to combine the benefits of reduced eddy current effects and higher flexibility. The



Fig. 5. Dependence of the piezomagnetic coefficient on magnetic field at various temperatures [2].

maximum ME sensitivity of the laminated composites is as high as over 3000 mV/cm-Oe at the resonance frequency of around 100 kHz. The ME effect increases with the volume fraction of T-D in the low concentration range due to the enhanced magnetostriction, but drops at high concentrations above the percolation threshold [101,151]. In a study on polymer-bonded cylindrical T-D/PZT ME MPC with various epoxy contents it emerged that the composite with an epoxy content of 14% weight fraction and 100–150 µm particle range exhibited better overall ME performance [152]. Jin et al. [153] showed dramatic enhancement of the ME effect in PVDF based composites by introducing chain-end cross-linking and polysilsesquioxane structures into the ferroelectric polymer films. It is believed that the structural modification leads to the formation of larger crystalline sizes and concurrent improvement in the polarization ordering and consequently better piezoelectricity. Recent research review showed the importance of considering the role of nano particles on the interface between organic and inorganic components [154]. Ourry et al. combined magnetic CoFe<sub>2</sub>O<sub>4</sub> nanoparticles of 6-nm and 9-nm sizes with PVDF by solution mixing [155]. The degree of crystallinity of neat PVDF was quite high whereas the presence of magnetic nano particles resulted in a decrease of the crystallinity likely because they prevent the growth of the PVDF crystallites. Gonçalves et al. [55] showed how the interface between CFO nanoparticles and PVDF composites can result in an optimized ME response. The addition of CFO nanoparticles into the polymeric spheres has almost no effect on the  $\beta$ -phase content, crystallinity or the onset degradation temperature of the polymer matrix. Multifunctional membranes comprised of CoFe<sub>2</sub>O<sub>4</sub> PVDF porous piezoelectric, magnetic, magnetostrictive and ME have been proposed [156]. The MCP with 1, 7 and 20 nanoparticle weight percent (wt%) were prepared by solvent casting and crystallization at room temperature, with thickness of ranging from 200 to 400 µm. A decrease is observed in the coupling coefficient which is attributed to the lower mechanical coupling between the nanoparticles and the polymer matrix when pores are present.

In addition to the interest in the magnetoelectric effect, the magnetodielectric effect (MDE) has also received recent interest and is related to the ability to control the dielectric permittivity as a response to applied magnetic fields. Materials exhibiting large MDE are suitable for new devices including radio frequency, tunable microwave, filters, four-state memories, magnetic sensors and spin-charge transducers [157–159]. Magneto-dielectric nanocomposite material based on nanoparticles with PVDF and PDMS was proposed for antenna applications [160,161]. Graphene-polyvinyl alcohol (PVA) magnetodielectric nanocomposite films with thickness 120  $\mu$ m were synthesized by solvent casting of PVA from a solution with dispersed graphene nanosheets [162]. The majority of work in this area has been on particulate composites that show how the dielectric response and magnetization is impacted by the volume content of the particles. The decrease in the magnetodielectric coefficient was related to the increase of dielectric losses and the deterioration of mechanical connectivity with increasing ferrite content and agglomeration [157,163–165]. Ferrite particles coated with a very thin layer of silica have been shown to prevent aggregation and thus increase its dispersibility showing strong ME coupling and enhanced ME sensitivity [166].

# 4. Modeling of magnetostrictive polymer composites

Modeling of the nonlinear hysteresis behavior, including the effect of compressive stresses, for monolithic magnetostrictive materials has been previously reported [167–171]. In an analogy to plasticity theory, Linnemann et al. [170] and Miehe et al. [172] considered that reversible and nonreversible components of the strain tensor and magnetic field components. Lu and Li [97] showed how nonlinear constitutive approaches can be used to study the thickness effects in T-D films. While extensive nonlinear hysteretic electromagnetic constitutive models have been developed, only limited micromechanics models with simplified microstructures have considered a nonlinear magnetostriction behavior. Guan et al. [173] used the Eshelby equivalent inclusion method taking demagnetization effects into consideration to estimate the average magnetostriction of composites. Hogea and Armstrong [174] considered rule of mixture (ROM) for predicting the magneto-viscoelastic response of composites comprised of magnetostrictive fibers. Homogenization models based on periodic unfolding, have been shown to yield three-dimensional magneto-electro-elastic periodic structures that can be extended to laminated magneto-electro-elastic composites [175]. The approach has been used to yield analytical relationships that can estimate the specific volume fraction where the magnetoelectric effective coefficient attain desired properties [176].

Analysis of magnetostrictive behavior for design at the current stage of development typically uses linearized approaches because of the simplicity of the approach. Linearized analysis starts by assuming a linear relation between the external magnetic field and the magnetic flux density in the material. When forming a composite with magnetostrictive phases and polymer phases, nonlinear constitutive models can be used. The nonlinear models for the polymer phases have been well established but only recently we have seen improved models for the nonlinear response of the magnetostrictive phases. A detailed approach that incorporates material stiffness, stress state, magnetic saturation and magnetization can be found in the work of Liu and Zheng [177].

#### 4.1. Micromechanics approaches

Periodic microstructure analysis using nonlinear micromechanics has been used to predict the effective behavior incorporating the impact of permeability of the polymer and magnetostrictive phases to determine the effective magnetization and in turn the behavior of the composite [178]. Micromechanics models have also been proposed by assuming a linear magneto-elastic response. Zhou et al. [179] used a mechanics of materials approach to predict the magnetostrictive behavior of T-D/epoxy and nickel/epoxy composites. The approach is simple and predicts the axial and transverse strains, however it does not directly account for the effects of the preload on the MPC. Altin et al. [58] used the upper bound rule of mixture (ROM) to determine the effective properties of MPC.

Homogenization approaches based on the Mori-Tanaka method and the asymptotic homogenization method have been also used to derive effective properties in MPCs [180–183]. For example, in three-phase magneto-electro-elastic fiber unidirectional reinforced composite with parallelogram cell symmetry [183]. ROM predictions on the elastic modulus show correlation to the 1–3 model [2]. The high-fidelity generalized method of cells (HFGMC) is a micromechanical method [184] for predicting the macroscopic response in periodic composite structures. Aboudi [185] used the HFGMC method to predict the homogenized electro-magneto-mechanical properties of similar composites using the nonlinear constitutive relations for the magnetostrictive phase proposed by Jin et al. [186]. Fig. 7 shows a modeling technique using actual microstructure images from scanning electron microscopy. In this modeling technique, a coupled finite element software is used to model the microstructure of the composite [187] (Fig. 6) using the constitutive response proposed in [177]. Multiphysics finite element software such as Comsol [188] allows for combining the constitutive response from various materials and also to include the interaction of strain with magnetic field in some of the phases. It therefore enables efficient analysis of MPC based devices.

The effective magnetostriction property in materials has been found to be dependent on the stress and temperature of the composite. Kim [182] predicted the effective properties of magneto-electro-thermo-elastic multilayer composites with good correlation to the Mori-Tanaka model. Currently available micromechanics models have been focused mainly on the linear coupling behavior. The morphology and the nonlinearities introduced in MPCs from the shape, size and distribution are not typically considered. The linear approach has been successful when the external stress and magnetic fields are low. Typical magnetostrictive materials, such as T-D, Galfenol, and amorphous Co<sub>77</sub>B<sub>23</sub> alloy, experience a nonlinear response when subjected to extreme conditions, either high mechanical stress and magnetic field or low stress and magnetic field.



Scanning Electron Microscope Image of Magnetostrictive Composite



Image Thresholding Identifying Terfenol-D (Black) and Epoxy (White) in MCM Model



Fig. 6. Simulations using multi-physics approaches showing scanning electron microscopy of MPC (a) conversion of this image to grayscale and then finite element mesh (b) the mechanical stresses post cure (c), and the distribution of the strain and magnetic field (d) [187].



Fig. 7. Magnetostrictive composite patches (with different geometry) (a) on a plate for sensing waves produced with a PZT actuator (b) [67].

#### 4.2. Modeling of MPC with magnetoelectric effects

Electrostrictive and magnetostrictive effects in 1–3 composites were examined using the framework of Landau phenomenological theory [189]. In [190] a phase field-type model is suggested and applied to particulate and layered ME composites using the coupled thermodynamic evolution equations with respect to polarization and magnetization, using the Eshelby's equivalent inclusion concept and the Mori-Tanaka method. Effective medium approximations were also used to study the ME coefficients of three-phase multiferroic particulate composites comprising magnetostrictive particles (T-D) and piezoelectric particles embedded in a conductive polymer matrix and compared with experimental results [191]. The  $\alpha$  values of these composites are highly related to complex permittivity, and the existence of a peak at a certain bias magnetic field related to the magnetostrictive, piezoelectric and elastic properties of the constituents.

The HFGMC approach can also be used to model the ME composites. The method incorporates piezoelectric constitutive relations for the magnetic, mechanical and electrical coupling equations. The mechanical deformations caused by the application of the magnetic field induce an electric field which together with the resulting magnetostriction can be used to model the MPC [192]. The results obtained from this model were compared with experimental results from T-D fiber/epoxy composites laminated to piezo-ceramics [193] and piezo ceramic rods embedded in a MPC [10].

The importance of understanding the effects of magnetic domains in the magnetostrictive phases has been also studied [194]. Computer simulations revealed the significant effects of magnetic domain structures on the ME responses of particulate composites with the longitudinal and transverse ME coefficients of 0–3 particulate composites with isotropic two-phase microstructures. Effects of inverse magnetostriction on ME coupling and magnetostriction saturation have been studied showing its possible influence on the ME effect [195].

Avakian et al. [196] proposed a nonlinear modeling approach of ME composites, where in the ferroelectric phase domain wall motion is taken into account, while the magnetostrictive constituents behave linearly. The constitutive equations are

implemented within a finite element (FE) framework to study particulate composites simulating the poling process and for the prediction of ME coupling and introducing influences of residual stresses and polarization scattering.

#### 4.3. Modeling elastomeric MPCs

The main focus in modeling elastomeric MPCs is generally directed at predicting the field dependent shear modulus. Most of considered approaches have been looking at continuum mechanics [48,197–200]. The classical Guth and Gold model can provide a good estimate of the mechanical behavior at low amounts of reinforcement but for higher values a percolation law is found to provide better results [48]. Zhang et al. [201] proposed an effective permeability model to predict the field-dependent shear modulus. At the cross-section normal to the columns, the composite is divided into various columns corresponding to the particles or the matrix. The effective permeability along the direction of columns is calculated using a parallel connection and rule of mixture approach based on the respective volume fraction of the constituents. In practice, the particles may be in contact in chain-like configurations which results in high local stresses and a large possibility of debonding. A Griffith type model has been proposed to study this behavior in first traction and it resulted to be able to predict the stress drop, as a consequence of the initial growth of the cavity. The model is also able predict the progressive drop of the slope associated with the extension of the debonded cavity [202]. Using the analogy of a sphere chain explains the Mullins effect and the impact of the bonding between the particles and its low impact on the stiffness when a magnetic field is present.

The magnetization-magnetic relations in elastomeric MPCs can also be modeled using a Langevin relation as proposed in [177]. The initial response can be approximated by dimensionless magnetization parameters in a Langevin function [49]. Han et al. [49] proposed a 1-D model with a few parameters to predict the magneto-mechanical coupling to be originated from the deformation-dependent magnetization (or permeability). The model was calibrated from the measurement of permeability –strain responses to predict both the stiffening effect and the magnetostrictive response.

Investigations have been also performed on how various organizational patterns of the particles may affect the behaviour of elastomeric MPCs [203,204]. A simple cubic lattice and body-centered model predicts a contraction of the elastomer with isotropic distribution of magnetic particles along the direction of a homogeneous magnetic field. It predicts that the Young's modulus decreases with an increase of the magnetic field. However, in contrast to the simple cubic lattice, the body-centered cubic lattice model provides an expansion of the MPC elastomers along the direction of a magnetic field. The hexagonal lattice model shows that materials with isotropic particle distribution contract along the external magnetic field, while its Young's modulus increases [203].

Applying Maxwell stress tensor with the magnetoelastic homogenization framework and the partial decoupling approximation, Galipeau and Castaneda [205] are able to model elastomers with ellipsoidal magnetic particles. They have showed the dependence of magnetic properties on the distribution, shape and concentration of particles. Similar to the modeling of magnetostrictive MPC, elastomeric MPC can be analyzed with micromechanics approaches. Sun et al. [206] have considered the interaction of Maxwell stress tensor at the boundary of magnetic particles with the rubber matrix. Accounting for the free energy from the rubber matrix, magnetic particles and dipole interaction with magnetic field, they have estimated the shear deformation in the representative volume element of the MPC for various magnetic field intensities, particle sizes and particle separation. This is more a boundary effect and magnetic dipole interaction among adjacent particles while magnetostriction is more complicated with domain rotation and expansion. Their results are consistent with predictions from methods based on mean field approaches.

#### 5. Applications of magnetostrictive polymer composites

#### 5.1. Sensing of ultrasonic waves

In guided wave methods (typically less than 100 kHz), mechanical stress waves are propagated along an elongated structure while guided by its boundaries allowing waves to travel over large distances. The method can be used to inspect pipelines over large distances and can also be used in inspecting long prismatic or plate-type structures. Polymer-bonded composites of Galfenol particles [67] have been employed in a magnetostrictive patch transducer systems that can be suitable for high frequency applications and at lower costs than other giant magnetostrictive materials like T-D (Fig. 7). In these experiments the composite patches (of different configurations) on a metallic plate were excited by a PZT actuator with an excitation frequency and amplitude of 60 kHz and 680 V peak-to-peak, respectively [207]. Flake based Galfenol MPC patches on the metallic plate had outstanding field sensitivity and successfully captured the induced waves. The sensor had the highest amplitude compared to the granular Galfenol or nickel MPC due to the large shape anisotropy of the Galfenol flakes [207]. The technique may have issues for implementation when applied to iron containing structures since it relies on picking up changes in the magnetic properties of the composite sensor. MPC offer increased advantages over monolithic magnetostrictive materials in sonar applications. The use of MPC can overcome some of the limitations of laminated systems to increase the bandwidth and stiffness properties to operate at frequencies greater than 1 kHz [60].

# 5.2. Actuators

A T-D strip bonded to a polymer band can serve as an actuator as discussed by Hong [208]. The center deflection of the laminate strip supported at its ends will act as an actuator when positioned near a permanent magnet. The strip reinforces the Terfenol-D and can result in multiple mm deformation. Coercivity can also be affected by the processing [81]. For motors, generators and sensors a lower coercivity is more desirable. For certain application, a long term memory may be useful which allows for accumulation of stress over time in which case a higher coercivity may be more desirable. Micro and nano-structuring can allow achieving these properties. Single-crystalline  $\alpha$ -Fe<sub>2</sub>O<sub>3</sub> in the form of hollow spheres and nano-cups with very large coercivity have been reported [209,210]. The properties are derived because of the complex shape anisotropy where the magnetic properties result from two-dimensional thin-film like walls of the three-dimensional nanostructures. Similarly, using hollow nano-spheres vs. bulk properties have also been found to significantly increase the coercivity by greater than 7 times [211]. The effects of increased polymer modulus results in a small decrease in the saturation elongation in magnetic ferrogels but it was found to affect the mechanical transition point corresponding to a given magnetic field [212]. In addition, for a given specimen length, there is an aspect ratio which will maximize the energy density and work that can be performed by the specimen [212].

# 5.3. Biomedical applications

Polymer based magnetic composites have attracted the attention of the tissue engineering community for membranes and tissue engineering applications [156,213–215], for drug delivery [216–218] and also for neural simulation [219]. There has been some concern with the agglomeration of nanoparticles and biological compatibility due to their reaction with biological membranes [214]. T-D/PVDF-TrFE composites have been used to provide mechanical and electrical stimuli to MC3T3-E1 pre-osteoblast cells. These new materials can potentially be used to mechanically and electrically effect body tissue material wirelessly to promote cell growth. Cell growth can be increased by more than twenty five percent when cultured under mechanical and electrical strain [142]. A flexible magnetic element can be an integrated artery or prosthesis suitable for wireless localized blood pressure monitoring. The sensor operating principle is based on the modulated scattering of electromagnetic waves by a magnetostrictive flexible strap with pressure detection, by means of magnetic permeability changes in the strap [220]. Researchers have shown how the size of pores can be controlled by using the interactions between the nanoparticle track-etched polyethylene terephthalate pores and further functionalization with poly(N-isopropylacryl amide) (PNIPAAm) [215]. The valve function of the MPC is achieved with an external control via a high frequency electromagnetic field demonstrated in water permeability experiments. Such devices can have potential application in drug release or mass separation applications. A recent example is a drug release system using a MPC with T-D and PLLA membrane containing a zeolite to release the ibuprofen drug. The application of an AC magnetic field results in a 30% increase in the release rate [218]. The magnetically driven release is believed to be due to swelling or erosion in the membrane induced by the magnetostrictive particles under the applied magnetic field. Zheng et al. also discussed the potential applications of nanostructured and nanoscale ME composites in bionics through recent demonstration of using ME composite nanoparticles for stimulating the brain [217]. ME MPC nanoparticles have been proposed to artificially stimulate the neural activity deep in the brain [221], Exploiting the difference in electric properties between normal and cancer cell membranes, ME particles have been shown to be capable of entering cancerous cells carrying a therapeutic payload and releasing the payload intracellularly with the application of an external magnetic field, while not affecting normal cells [219]. This recent demonstration shows the potential use of MPC for these types of biomedical applications.

#### 5.4. Sensors for magnetic fields and electrical currents

Several researchers have demonstrated that a fiber Bragg grating (FBG) can be bonded onto a monolithic T-D magnetostrictive alloy to form a magnetic or electric field sensor [222-224]. MPC with epoxy-bonded T-D particle coupled with FBG as a strain sensor has been proposed for magnetic field measurement [225,226]. When a magnetic field is applied, the magnetostrictive strain from the MPC will be directly coupled to the FBG thereby shifting the peak reflection of the FBG to a longer wavelength. The MPC FBG sensor not only possesses a large quasi-static peak wavelength shift of 0.68 nm at an applied magnetic field of 146 kA/m but also solves the issues with the bulky monolithic counterparts [225]. The dynamic also had a ten-fold sensitivity increase at the fundamental shape resonance. A current sensor device with an optical fiber coupled with a strain distribution converter has been proposed. In this implementation, a MPC associated with the strain distribution converter acts such that a change in shape of the magnetostrictive material modulates the distribution of the FBG grating periods [227,228]. Such an optical current sensor with a wider dynamic range can be realized with a graded particle size distribution converting the magnetostrictive strain into frequency chirping in proportional to magnetic field [229]. In another fibre-specific application, a fiber Bragg Grating sensor is used for DC and AC magnetic field measurements by coating the fibre with a thick layer of MPC (30% volume fraction) [230]. The sensor was successfully tested in magnetic fields of up to 0.750 T under static conditions. Polymer composites suitable to be used as magnetic field sensors for advanced applications have also been proposed by using  $\delta$ -FeO(OH) nanosheets inside a piezoelectric P(VDF-TrFE) polymer matrix<sup>72</sup>].

Magnetostriction and quantum tunneling have also been combined for measuring the current induced by strain with increased sensitivity and a high gage factor of approximately 80 [231]. In this approach, the strain is transferred using mechanical coupling between the magnetostrictive phase and the nanoparticles. The strain results in changing the spaces between the nanoparticles thus leading to the quantum tunneling effect. Minor changes in the spacing result in exponential tunneling current being produced.

#### 5.5. Vibration isolation and active control

The increase in the stress and stiffness level of composite elastomers under a magnetic field lays the basis for their potential application for an active control of stiffness and vibration of structures [47]. A shear mode vibration isolation system using the MR properties of elastomer-based MPCs have been proposed [232]. The system is comprised of a dynamic mass, static mass and smart spring elements made with a carbonyl iron reinforced rubber elastomer. The adaptive tuned vibration absorber can have its modulus controlled by an applied magnetic field. The resonance frequency of the system can be varied from 27.5 Hz at 0 A up to 40 Hz at 0.5 A.

Vibration suppression using elastomers [233,234] and Ni-Mn-Ga based polyurethane MPC [85] has also been investigated. The acoustic attenuation in Ni-Mn-Ga/polurethane composite have been reported to be ideal for systems requiring mechanical energy dissipation [85]. Resonances beyond the principal mode are highly damped in the Ni-Mn-Ga samples with the transmitted amplitude stress in the frequency range above 550 Hz path leads to destructively interfere with each other resulting in a slowly decreasing trend.

#### 5.6. Magneto-electric sensors and devices

This class of magneto-electric composites generally refers to materials able to demonstrating ferroelectricity and ferromagnetism and has been the subject of several review papers in the literature in previous years [9–13]. The coupling between the magnetostrictive phase and piezoelectric phase in the composite leads to a giant magnetoelectric response that can be deployed to variety of sensors and devices. ME laminate composites are mostly fabricated by bonding a magnetostrictive material to a piezoelectric macro fiber composite or laminate. A larger ME effect (greater than 10x) can be obtained in composites consisting of two functional phases: a magnetostrictive phase, in which a strain is produced by application of a magnetic field and a piezoelectric phase, in which a change in electric polarization is produced by an applied stress [235].

In the absence of a piezoelectric phase, research shows that the pure polyurethane shows lower current values than ones filled with magnetostrictive particles [84]. The magnitude of magnetoelectric current is independent of the applied dc bias magnetic field and is a linear in function of the ac alternative field or applied frequency. In some cases lower magnetostric-tive materials have shown a higher ME which may indicate that the magnetostrictive property does not have a direct effect on the ME effect.

The ME material has many applications including optical wave modulation, data storage and switching, spin-wave generation, amplification and frequency generation [236]. In a 3-phase composites it is possible to use the magnetostrictive effect in the magnetostrictive phase and the piezoelectric effect in a piezo-electric phase. When a magnetic field is applied to such a composite, it induces a strain in the magnetostrictive phase. That strain is then transferred into the piezoelectric phase to generate a piezoelectric voltage or charge proportional to the applied magnetic field [237].

Wan et al. [238] proposed a ME composite structure which combined the T-D/epoxy (in 0–3 form) and PZT/ epoxy composite operating by the longitudinal vibration driving the piezoelectric output. The ME MPC provides a giant magnetoelectric voltage coefficient at the fundamental longitudinal resonance mode of the magnetostrictive phase with low eddy current loss. The performance in these composites is limited by mechanical shear lag and demagnetization. The stress nonuniformity and significant strain decay near the ends imply that the previous models are only accurate for the ME sample with very large aspect ratios [239]. Ryu et al. achieved a ME voltage coefficient up to 4.68 V/cm-Oe at 1 kHz using PZT and T-D plates [240]. The polymer layer between the PZT phases and magnetostrictive material also affects the mechanical coupling in a negative way. Using low temperature jet vapor solder bonded (<125 °C) magnetoelectric composite fibers and room temperature curing epoxy has also been reported with improved energy harvesting capabilities [241]. A 3 phase composite with metal, ceramic and PVDF showed a coefficient exceeding 9 mV/(cm-Oe) [146]. Another system showed a maximum of magnetoelectric coefficient of 82.58 mV/cm-Oe obtained in an inducing magnetic field  $H_i$  = 200 Oe and with a bias magnetic field H<sub>dc</sub> = 1000 Oe [145]. Cellulose-based magnetoelectric laminate composite that produces considerable magnetoelectric coefficients of  $\approx$ 1.5 V/cm-Oe have also been reported comprising a Fano resonance that is ubiquitous in the field of physics, such as photonics devices [242]. Such a phenomenon is the manifestation of interactions among weakly coupled harmonic oscillators which have different but close resonant frequencies and are various states of resonant transmission and resonant reflection for certain frequency ranges.

# 5.7. Stress sensing and health monitoring

Stress sensing in MPC typically uses the Villari effect, which is a change in the magnetic susceptibility of a material when it is placed under mechanical stress [6,243]. This phenomenon can be the basis for a sensing mechanism that determines the stress in a structure by monitoring the magnetic susceptibility of an attached or in-situ MPC sensor. Table 4 shows the

# Table 4

Summary of MPC used for stress sensing applications.

Magnetostrictive material	Polymer	Stress range	Sensor	Mechanical properties	Ref
CoO-Fe <sub>2</sub> O <sub>3</sub>	Phenol	0-30 MPa	Inductance	E = 50 GPa	[81]
T-D	Epoxy	0-30 MPa	Hall Probe	-	[92]
T-D	Viny-Ester	0-13 MPa	Gauss meter	1.3 GPa	[248]
T-D	Ероху	0-60 MPa	Inductance	-	[247]

typical stress ranges explored using MPC reported in the literature. Compared to their monolithic counterparts MPC stress sensors have lower cost (less use of rare earth materials), ease of manufacturing, handling, and possibility of making complex shapes [58]. MPCs were included in a composite beam with woven materials under bending load [244]. The results showed that a clear relationship can be established between the magnetic flux density measured at the surface and the stress state inside the beam. Chen and Anjanappa [245] introduced MPC layers with thicknesses ranging from 100 to 300 µm within carbon fiber laminated composites with the intention of providing health monitoring of composite structures. A sensing device based on a U-shaped magnetic and sensing coil wound on the two arms of the core was realized. The sine function magnetic field is applied to the magnetostrictive film showing a voltage change when a delamination is present. Concentrations of particles that are smaller results in a larger homogenous distribution of particles [91]. This results in higher magnetic flux density changes and better sensitivity for using MPC for applications based on the Villari effect [92]. Haile et al. [246] also investigated the feasibility of using T-D particles within a carbon-fiber AS4 and 3501-6 epoxy resin material system to sense early signs of fatigue damage. The sensing approach is based on the notion that magnetostrictive particles will undergo irreversible changes in magnetization intensity when subjected to cyclic loading, and that this change can be captured with an induction coil sensor. The results indicate changes in the pick-up coil measurements that correlate with fatigue cycles. However, the approach suffers from the drawback of possibly reducing the strength of the composite, power requirements for drive coil and possibly a low signal below the noise floor [246]. Stress sensing is based on inductance measurements by placing a sensor in an air gap near the material. The approach was proposed by Al-Hajjeh et al. [247] to measure the inductance in an MPC bonded to an aluminum substrate at various angles to the applied load (see Fig. 8). The setup was able to provide accurate wireless measurement of the magnetostrictive material. The authors were able to reduce the impact of the shape and improved the performance by measurements up to 900 microstrains [247]. MPC with rectangular Co ferrite particles bonded by a phenol polymer [81] have been used in stress sensing applications. The interesting feature for stress sensing when using these particles is that most of the magnetostriction can be realized between the 50 and 130 kA/m range.

The polymer will reduce the global constant but it allows for sensing over a wider stress range. It is also likely to impact the temperature dependence of the material as well since the permeability was found to change dramatically with an increase in temperature under tensile stress application.

A T-D/polyurethane coating added to the kevlar fabric coated fabric targets showed significant damage reduction by absorbing the energy from impact [86]. Lower performance due to damping and higher stiffness material may be the reason for the response observed. The tension stress in the fabric caused by the T-D magnetostriction may impact the ballistic behavior by encouraging more damage to occur. It is also possible to tag or include the magnetostrictive material in critical places. In one study the magnetostrictive particles were applied only in the outer layer of structure and only in the mid-span region of a beam type specimen [248]. This concept allows tailoring of the magnetostrictive tagging and results in considerable cost saving and improved application efficiency. Monitoring can be achieved by measuring the B versus H curve with an excitation coil and a pick up coil. The method is based on mutual coupling between 2 coils, i.e. based on mutual



Fig. 8. Stress sensing on MPC composite using a toroidal coil [247].

inductance [81]. Trovillion et al. [249] studied the magnetic characteristics of neat resin and glass-fiber reinforced magnetostrictive composites subjected to axial load. Gaussmeter probe and small integrated circuit Hall effect devices were used to measure the magnetic field and the results were comparable up to 2 kN in the composite.

#### 6. Concluding remarks

Considering the significant amount of research conducted in the past two decades and summarized in this review, the authors of this review believe that future advancements are being limited by the complexity of this problem at numerous levels. The difficulty in conducting representative experiments is limiting the ability to develop advanced modeling approaches. In addition, we address below specific recommendations and concerns raised by us and other researchers in the field.

# 6.1. Fundamental research

Starting at the micromechanical level, it is important to understand the forces that determine the order of the particles in the matrix. Time, strain amplitude, frequency, magnetic field, shear rate, and temperature are expected to influence the forces ordering the particles and have not yet been studied systematically. Secondly, a unified understanding of the interaction between the electromagnetic behavior, mechanical loads and thermal conditions is also needed. How these boundaries interact with the effects of microstructural morphologies on the overall response of MPCs in sensor applications is currently not adequately addressed in the literature. Further, most of the applications using MPC materials are subjected to combined loadings for example compression/shear or tension/shear whereas the research on combined loading has been very limited and there is little understanding if the research conducted can address the needs of these applications. In dynamic applications, the force, frequency, strain, magnetic field strength and uniformity as well as the directions between the load, magnetic field and particle orientation are all areas that need further research. Research publications need to better document their measurement approaches so comparative analysis can be more easily made. In addition, while there is growing understanding of the reinforcement effects on the response, there is a limited understanding of the trade-offs involved when considering a lower stiffness matrix for example or what the effects of the fatigue response when the composite is made more brittle with a higher percentage of reinforcements.

The nonlinear magnetostrictive response in MPCs is greatly influenced by the residual stresses in the material and any externally applied mechanical loads. The response is also affected by the temperature changes in the magnetostrictive phases of the MPCs due to the flow of eddy currents at high frequencies. Even though these eddy currents are greatly reduced by the polymer binder in MPCs compared with the monolithic materials, they can still generate significant heating under high frequency (at 100's of KHz) magnetic fields, increasing the body temperature of MPCs. These temperature changes could be detrimental to the viscoelastic polymers, thus degrading the performance of the sensor. Dynamic response of inverse magnetostriction in MPC has not been significantly investigated. Similarly, MPCs prepared by using fibril type reinforcements have not been adequately evaluated for dynamic effects. In the absence of a piezoelectric phase, the research shows that the pure polyurethane shows lower capabilities than ones filled with magnetostrictive particles. The effect of volume fraction and the percolation value should be studied on the magnetoelectric current.

#### 6.2. Magnetostrictive reinforcement

T-D microspheres may provide more promising results as nickel based microspheres have shown positive results [53]. Some work on T-D microspheres have been presented using a spark-erosion process [53]. Oxidative effects have been studied on elastomers [126] but there has not been a systematic study of oxidative effects on MCP. The low k<sub>33</sub> results from the modulus value of the epoxy. A higher stiffness polymer will increase the coupling behavior. Based upon the previous results, ball-milling [66] offers a cost-effective pathway towards realizing large volumes of FeGa alloys having moderate values of magnetostriction. For the ME effect, there is a need to further explore the effect of the magnetostrictive mechanism acting on the piezoelectric response of nanofiber nanostructured composites [217]. Piezoresponse Force Microscopy has been recommended as a tool to study the magnetostrictive effects at the nanoscale especially together with more studies conducted for in vivo applications [217]. Research is needed to understand the effects of controlling the magnetic domain structures in the MPC via internal residual stress engineering. For example, as suggested by Ma et al. [194], to co-sinter the material under unaxial or biaxial stresses. The approach can help to obtain better properties and to get rid of the bias field used when applying these materials as has been seen in other non-polymer based systems [250].

# 6.3. Interface effects

The internal cracking or interface degradation may lead to reduced cyclic magnetostriction as the strain transfer between particles and matrix within the composite is strongly reduced. This is an area that has not been particularly studied particularly that increasing the particle content and interface contact by optimization of the processing. The magnetostriction hysteresis in MPC, especially the elastomeric ones, is disadvantageous and methods for improving the interface should be investigated, such as chemical purifications of the surface to provide better adhesion with the silicone [107]. The CPVC

approach [122] may need to be considered. Perhaps measurement of porosity in the powder before filling with polymer is a parameter that needs to be reported by the researchers since it gives an idea of the space between the particles. Statistical analysis on static and dynamic T-D properties have been reported [24], but analysis of other reinforcements is still sparse. Further research on the mechanics of magnetostriction in magnetostrictive nanowires analogous in the research directions in single chain magnets or magnetic nanowires [251,252] are needed to understand how they can be applied to MPC in structural network formations. Application of these materials will require an accurate understanding of material properties under magnetic field bias, mechanical preloads as well as different AC fields and various excitation frequencies.

# 6.4. Modeling

The overwhelming efforts in modeling the constitutive response shows significant emphasis on T-D behaviors but it is important to consider the other magnetostrictive materials used in MPC. Typical magnetostrictive materials, such as T-D, Galfenol, and amorphous Co<sub>77</sub>B<sub>23</sub> alloy, experience a nonlinear response when subjected to extreme conditions, either high mechanical stress and a magnetic field or low stress and a magnetic field. A unified model for magnetostrictive materials that includes magnetic effects, preload stress under extreme conditions has not yet been discussed in detail and primarily because MPC have not been tested under extreme environments. Furthermore, models that are capable energy dissipation are need for modeling advanced magnetostrictive materials and their composites used in the dynamic regime. In fact, it appears to be urgently necessary to accurately predict the nonlinear and hysteretic electro-magnetic response of MPCs. Limited amount of work has been observed in the literature on using modeling techniques for assessing the performance of MPC based devices. Future research and studies are needed in order to benchmark and assess the various modeling approaches in the literature. There is significant room for multi-scale methods within the multi-physics analysis frameworks currently available. One possible strategy would be to embedd the micromechanics based HFGMC approach discussed above into a finite element model where the global analysis is performed using a finite element approach and the micromechanics approach is evaluated at the integration points.

#### 6.5. Future applications

The use of nanoscaled T-D particles is a promising step forward particulary if domains can be arranged at the smaller particle sizes. A higher sensitivity of these sensor particles might result in a reduced particle content, also creating the opportunity to reduce the total weight of the composite. When applying MPC in laminated composites for structural health monitoring and indicator for fatigue damage, researchers should consider how to transfer the stress from the laminate into the MPC. Increasing laminated composite material reliability should be achieved without reduction of the mechanical properties. Consideration may be given to low-modulus materials that can blend easier within the matrix so it can induce less stress concentrations. Also, locating the pick-up coil away from the specimen and power-requirements for the drive coil are all practical considerations that need to be assessed. For stress sensing applications it is advisable to consider the best modality for sensing the change in stress with a combination of hall sensors or inductance meters. The use of coupled methodologies to sense the changes in the magnetic phases of the composite may be an area deserving new attention in the literature.

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