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Static and Dynamic Magneto-Elastic Sensing Properties of Fe-Al Alloy Powder-Epoxy Composite Patches

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Abstract: By combining the two types of magnetoelastic and magnetochromatic materials in an epoxy, we can make a hybrid system that exhibits an optical response due to an elastic strain. It could be used in structural health monitoring, for real-time monitoring of crack propagation or general evaluation of the condition of a structure, both visualized by a change in color. In this study, magnetostrictive polymer composites (MPCs) with Fe_{81}Al_{19} (Alfenol) alloy particles are evaluated to determine magneto-elastic properties in composite patches attached to a surface, prior to understanding the full hybrid magneto–elasto–optical interactions. To measure static magneto-elastic performance, a tension apparatus within a solenoid was fabricated to apply uniform strain to the MPC patch on an aluminum dog-bone substrate. It was demonstrated that, for epoxies with an elastic modulus higher than ~0.1 GPa, a tensile strain/stress applied to the composite improved magneto-elastic coupling, resulting in increased permeability values, at least up to strains of 0.1%. Composites were fabricated with both spherical and flake-shaped powders, with flake-shaped powders exhibiting better magnetic responses than those with spherical morphology. Alfenol MPCs were also measured dynamically at ultrasonic frequencies, exhibiting comparable dynamic sensing performance to Galfenol at 120 kHz using ultrasonic guided wave techniques.

Keywords: magnetostriction; metal-polymer composite; Fe-Al alloy; structural health monitoring; magneto-elastic coupling

1. Introduction

1.1. Motivation

Many structural health monitoring (SHM) solutions observe only discrete points of strain or require processing of an indirect acoustic signal to understand what is happening in the structure beneath. It is particularly challenging to apply SHM to ships and aircraft, due to their large and varied physical size and geometry, extreme operational and environmental conditions, and limited sensing technologies available for SHM [1]. Multifunctional materials, however, could be used in a surface coating composed of functional powders inside of an epoxy matrix and provide additional SHM solutions. However, there are many challenges in adapting such solid-state solutions to a large-scale coating, including powder properties, functional physics interactions and powder–matrix interactions. Nevertheless, hybrid magneto–elasto–optical systems have the potential to translate surface strain into a color change for SHM, mediated by the magnetic coupling between two different functional materials, specifically magnetostrictive and magnetochromatic materials.

1.2. Background of a Hybrid System

The relationships between magnetic, elastic and optical properties are illustrated in Figure 1 as the interaction flow from magnetoelastic (strain-to-magnetization) to magneto-optic (magnetization-to-tunable color) interactions. In magneto-elastic materials, there is a
shape, dimension or elastic properties change in the presence of magnetic fields, and vice versa. This can be used either directly for actuation or the reverse effect allows for sensing and energy harvesting. The inverse magnetostrictive effect, or Villari effect, can then be used to trigger a color change in the magnetochromatic (MC) material, due to localized stray magnetic fields surrounding the strained magnetostrictive (MS) particle. Fe-based MS materials can be used in many different forms: lamination, strips, sheet and powders. Fe-Ga (Galfenol) and Fe-Al (Alfenol) alloys possess a unique combination of good magnetic properties along with good mechanical properties [2,3]. Compared to other magneto-elastic materials, they have very low saturation fields of ~100 Oe, very low hysteresis and limited dependence of magnetomechanical properties at temperatures between 253 K and 353 K [4]. Thus, Galfenol and Alfenol rolled-sheets can be used in a laminated composite with PZT and carbon fibers for energy harvesting and cantilever-type sensors, respectively [5,6]. We have contributed by developing highly textured Galfenol and Alfenol sheets exhibiting single-crystal-like growth through thermo-mechanical processing [7–9].

In contrast to magneto-elastic materials, magnetochromatic (MC) materials reflect specific wavelengths of light when exposed to external magnetic fields. Colloidal magnetic crystals have attracted increasing attention since MC microspheres were developed in 2009 by a research team led by Professor Yin at the University of California, Riverside [10]. The MC microspheres were prepared through a simultaneous magnetic assembly and UV curing process in an emulsion system. Superparamagnetic Fe₃O₄@SiO₂ colloidal particles were self-organized into ordered structures inside emulsion droplets of UV-curable resin. They demonstrated the excellent capability of fast on/off switching of the diffraction by changing the orientation of the crystal lattice relative to the incident light using magnetic fields, being applicable to color displays, rewritable signage and sensors.

1.3. Magnetostrictive Polymer Composite Systems

This hybrid system has the potential to be applied as a paint, but small-scale particles are needed to allow the particles to remain suspended in the solution. The small-scale particle interactions and the interactions of these particles in an epoxy need to be characterized before developing the paint. After understanding the various levels of interaction in such a hybrid system, it may be possible to use this hybrid paint for SHM, enabling real-time evaluation of the condition of the structure and monitoring of any defect of the structure. Magnetostrictive polymer composites with Terfenol-D powders have been preferred for use in actuation to realize high strain and energy density at low frequencies, while Fe-Ga alloy (Galfenol) powders have been considered for use in magnetoroheological (MR) fluids, MR elastomers for tactile sensors and SHM systems as composite patches to detect ultrasonic guided waves [11–15]. Fe-Al alloys (Alfenol) also have good magneto-elastic coupling coefficients similar to Galfenol, although the peak magnetostriction value is only a little higher than half that of Galfenol [2]. While exhibiting lower strain values, aluminum additions are also a more cost-effective option, with Al being less than 1% of the cost of gallium [16]. In this study, the magneto-elastic coupling properties of polymer-matrix composites with magnetostriuctive Alfenol particles are investigated as a preliminary work for understanding the hybrid interactions of magnetostrictive and magnetochromatic particles in an epoxy. The performance of the composite is directly linked with the strain transfer from the substrate to the epoxy, and the epoxy to the functional particles. Therefore, a more detailed study is essential, as the modulus of the composite and its ability to transfer strain are linked to the powder morphology, powder weight percent and the stiffness of the epoxy matrix. We also demonstrate the ultrasonic guided wave sensing performance of magnetostrictive polymer composites (MPCs) with Galfenol and Alfenol powders. Alfenol MPCs with flake-type powders showed notable static and dynamic sensing performances.
was determined by scanning electron microscopy (SEM), as shown in Figure 2. After ball-milling, the rounded and spherical atomized powders turned into flake-type particles was determined by scanning electron microscopy (SEM), as shown in Figure 2.

Two different powder morphologies (spherical and flake) of Alfenol were used for preparing magnetostrictive polymer composites (MPCs). Atomized powders with a spherical morphology were produced by American Elements. These powders were then wet ball-milled using ethanol with a SPEX 8000 mixer/mill to obtain flake-type powders. A set of sieves was used to obtain Alfenol powders with particle diameters in the ranges of 53–106 and 106–150 μm. The powder morphology of the 106–150 μm diameter particles was determined by scanning electron microscopy (SEM), as shown in Figure 2. After ball-milling, the rounded and spherical atomized powders turned into flake-shaped powder. The atomized powders are additionally polycrystalline, as shown in the inset of a spherical powder image of Figure 2.

**Figure 1.** Schematic diagram of relationships between magnetic, elastic and optical properties, and the interaction flow from magnetoelastic (strain → magnetization) to magneto-optic (magnetization → tunable color due to wavelength change [17]) interactions along the filled circles and red thick arrows, where the outer corners represent external stimuli and the inner corners are the responses.

### 2. Materials and Methods

2.1. Static Test Setup

Figure 2 shows the composite disc sample preparation process using Fe$_{81}$Al$_{19}$ (Alfenol) powders and epoxy for basic characterization of physical, magnetic and magnetoelastic properties. Two different powder morphologies (spherical and flake) of Alfenol were used for preparing magnetostrictive polymer composites (MPCs). Atomized powders with a spherical morphology were produced by American Elements. These powders were then wet ball-milled using ethanol with a SPEX 8000 mixer/mill to obtain flake-type powders. A set of sieves was used to obtain Alfenol powders with particle diameters in the ranges of 53–106 and 106–150 μm. The powder morphology of the 106–150 μm diameter particles was determined by scanning electron microscopy (SEM), as shown in Figure 2. After ball-milling, the rounded and spherical atomized powders turned into flake-shaped powder. The atomized powders are additionally polycrystalline, as shown in the inset of a spherical powder image of Figure 2.

**Figure 2.** Powder morphology and composite disc sample preparation for static characterization of physical, magnetic and magnetoelastic properties.
Powders were mixed with water clear polyurethane (WC-756 A/B, BJB Enterprises) with weight percentages of 25–82%, then poured into a 32 mm × 32 mm mold. The composite was cured at RT for 24 h, then cut into discs with a diameter of ~6.4 mm and thickness of ~1.4–1.8 mm. Magnetization (M-H) and magnetic flux density (B-H) were measured as a function of applied magnetic field at room temperature (RT) using a Lakeshore vibrating sample magnetometer (VSM) under applied magnetic fields up to ±20 kOe; applied field values were corrected for sample shape anisotropy. While the shape of the particle should be used if the particles are decoupled, here the geometry of the sample was used; more analysis will be needed to determine the exchange coupling between particles and the effect of their individual geometries. Additionally, a polymeric c-clamp was used to apply compressive stress to the composite discs during the VSM measurements, to determine the change in permeability with compressive stress; the stress was applied in the direction of the sample thickness and the magnetic field was applied in the plane of the disc (see the right lower image in Figure 2).

A tension apparatus was designed and fabricated to measure magnetic response while applying strain to the composite patches. Composite samples were prepared using three different polyurethane types with varying stiffness (BJB Enterprises WC540, WC756, and WC792), and then cut into rectangles of dimension 10 mm (width) × 24 mm (length) × 1.6–2.0 mm (thickness). These were mounted on a dog-bone aluminum substrate using cyanoacrylate (i.e., Super Glue) and placed in a solenoid coil as shown in Figure 3. A strain gauge was attached to the opposite side of the dog-bone specimen (directly to the aluminum) for monitoring the strain up to ~1000 parts per million (ppm) during the magnetic response measurement. Magnetization was measured by the pickup coil around the composite sample, and the magnetic flux density coming out from the composite was detected by the hall sensor next to the composite. The gauss probe measures environmental magnetic field for reference. The measurement system consists of signal generator, power amplifier, DC power supply, flux meter, strain indicator, Gauss meter and oscilloscope. A signal generator (33210A, Keysight Technologies, Santa Rosa, CA, USA) generates a sinusoidal signal to a linear power amplifier (7224, AE Technon, Elkhart, IN, USA), and it drives the magnetic driver to generate magnetic field through the specimen. An analog DC power supply was used for current input to the hall sensor (FH-301-040, Meggitt, Coventry, UK) and a Vishay 3210 signal conditioning amplifier was used to amplify the signal from the Hall sensor. A flux meter (MF-10D, Walker LDJ Scientific, Worcester, MA, USA) was used for integrating signal from the pickup coil. A strain indicator (3800, Vishay Instruments Division, Columbia, MD, USA) was used to display the strain of the aluminum dog-bone sample mount. A Gauss meter (450, LakeShore, Carson, CA, USA) was used to measure the environmental magnetic field inside of the magnetic driver. The sensor response was captured by a digital oscilloscope (InfiniiVision DSOX3014A, Keysight Technologies).

![Figure 3. Tension apparatus setup with a dog-bone aluminum substrate and solenoid coil to generate the magnetic field.](image)

### 2.2. Dynamic (Ultrasonic) Test Setup

To evaluate performance at higher frequencies, a guided wave test was performed. Flakes of Alfenol and Galfenol (powder diameters of 106–150 µm) were mixed with an epoxy resin separately (105 Epoxy Resin/209 Extra Slow Hardener, West System, Bay City,
Three powder-epoxy composite samples (i.e., MPC patches) were prepared for each alloy with a powder weight fraction of 50%. The wet composite mixture was spread in a 0.635 mm thick 3-D printed stencil, which was attached to a test plate specimen. The 3-D printed stencil has a unidirectional shape, which could improve magneto-elastic coupling performance through magnetic shape anisotropy and also shows a different directional response compared with the magnetostrictive composites. Alfenol and Galfenol composite patches were directly applied to a circular 2024-T3 aluminum plate, with dimensions of Ø610 mm × 1.02 mm in thickness. Three magnetostrictive composite patches with dimensions of Ø25.4 mm × 0.635 mm for each alloy were applied 8 inches from the center of the plate at ±15° intervals as shown in Figure 4. The composite patches were aligned such that their easy/long axis was oriented toward the center of the plate. A 0.635 mm diameter Lead zirconate titanate (PZT) disc (Piezo Systems, Inc., Cambridge, MA, USA) was used as a guided lamb wave (GLW) actuation source, attached at the center of the plate. A custom-made magnetic circuit device containing an annular sensing coil and a cylindrical biasing magnet was employed to detect the change in magnetic properties induced by the elastic wave propagating through the composite patch. Tape was applied as a damping material to reduce the GLW reflections from the edge boundary of the circular plate in order to effectively investigate the incident GLW mode arriving directly from the elastic wave source. Figure 4 shows the experimental setups for the GLW tests.

Figure 4. Experimental setup for ultrasonic guided wave evaluation using Galfenol and Alfenol particle epoxy composites with a powder weight fraction of 50% and particle sizes of 106–150 µm.

3. Results

3.1. Static Magneto-Elastic Sensing Properties

3.1.1. Physical and Magnetic Properties of Composites

Figure 5 shows the trend of the density (ρ), the saturation magnetization (4πMS) and the initial/maximum permeability (μi and μmax.) as a function of powder weight fraction (WP) for the powder size of 53–106 µm. For both particle types, the magnitude of ρ increases with increasing WP because ρ for the Alfenol powder (7.11 g/cm³) is higher than that of the polymer (1.07 g/cm³ for WC756 in Table 1). The composites with spherical powders exhibit higher ρ and 4πMS than those with the flake-type powders over the entire range of WP. The high viscosity (650 cps at RT) and relatively short solidification time (15 min) of the epoxy affects the ability of randomly aligned flakes to stack efficiently, resulting in an increase of volume even at the same weight fraction compared with the sphere powders. The general trend of the 4πMS and ρ data is similar between particle morphologies and is in close agreement comparing the same morphology. However, the relative shape of μi and μmax. with increasing powder weight percent is quite different between flake and sphere. In the case of sphere, the permeability monotonically increases with an increase of WP, showing maximum values of 6.3 and 6.8 at WP of 81 wt.% for μi and μmax., respectively. By
contrast, the composites with flakes exhibit drastic changes in $\mu_i$ and $\mu_{\text{max}}$ as we increase the weight fraction of powders, and the maximum value reached 65 and 109 at $W_P$ of 75 wt.%, respectively. Comparing with an Alfenol disc, which was prepared by the arc-melting technique, the maximum permeability of the composites is almost 67% of the bulk sample result (96 and 163 for $\mu_i$ and $\mu_{\text{max}}$). It is possible that the alignment of the flake particles would improve along the in-plane direction at the higher weight percentages. Beyond the potential ability for the flake particles to stack, the demagnetizing factor due to the particle shape also has an important role in applications, where it is critical to achieve a high magnetic response at a low magnetic field. Spherical powders have a low field sensitivity at low magnetic fields due to their high demagnetizing factor. Here, the demagnetizing field of spheres is about ~6 kOe (equivalent to one third of $4\pi M_S$). Therefore, flake powder has an advantage for enhanced magnetic sensing performance at low applied magnetic fields.

![Figure 5. Physical and magnetic properties](image)

**Figure 5.** Physical and magnetic properties ((a) density, (b) saturation magnetization, (c) initial permeability, and (d) maximum permeability) of Alfenol epoxy composites with the powder sizes of 53–106 μm as a function of powder weight fraction.

**Table 1.** A summary of Alfenol flake-epoxy composite samples for static tensile test on a dog-bone aluminum substrate.

| Sample ID | Powder Weight/Volume Fraction ($W_P/V_P$) | Particle Size (μm) | Young's Modulus ($E_C$) | Product Name * | Density (g/cm³) | Young's Modulus ($E_E$) |
|-----------|------------------------------------------|--------------------|-------------------------|----------------|----------------|-------------------------|
| C1        | 0.417/0.0964                             | 53–106             | 0.0564 GPa              | WC540          | 1.06           | -                       |
| C2        | 0.521/0.1407                             | 106–150            | 0.5042 GPa              | WC756          | 1.07           | 0.098 GPa               |
| C3        | 0.537/0.1521                             | 53–106             | 10.7263 GPa             | WC792          | 1.10           | 1.86 GPa                |
| C4        | 0.589/0.1818                             | 106–150            | 14.0480 GPa             | WC792          | 1.10           | 1.86 GPa                |

* BJB Enterprises.

In order to verify the transfer of strain through the epoxy to the powders, $M−H$ curves for the disc-shaped composite samples were measured under various compressive stresses. The stress was applied in the thickness direction and the $M−H$ curve was measured along the in-plane direction of the disc. Prior to applying them to stress, there was a comparison between sphere and flake particles in magnetization and permeability in the composites with powder weight fractions of 60.8% for flake powders and 76.3% for
spherical powders, as shown in Figure 6a,b. The flake-based epoxy shows better field sensitivity than the sphere-based epoxy at the low field ranges; the sphere epoxy only has a higher saturation magnetization due to the higher weight fraction. The magnetic hysteresis for both samples was very low, while still exhibiting the non-linear magnetization responses expected from ferromagnetic particles. The permeability and permeability change linearly increased with an increase of out-of-plane compressive stress, as shown in Figure 6c,d. The permeability change for the flake sample shows a higher response than the sphere sample, which indicates a larger response due to applied stress, and a greater level of magneto-elastic coupling.

![Figure 6. Comparison of (a) B-H curves and (b) permeability of Alfenol epoxy composites using flake powder (60.8% weight fraction) and spherical powder (76.8% weight fraction) with a powder size of 53–106 µm. Comparison of compressive stress dependence of (c) initial and maximum permeability and (d) permeability change between the flake and spherical powders.](image)

3.1.2. Tensile Test of Composites

To evaluate tensile performance more quantitatively, rectangular-shaped composites were attached to the center of gauge section of dog-bone tensile specimens. Table 1 summarizes the composite samples for the tensile test. The tension apparatus was designed to control elongation of the aluminum dog-bone specimen within the elastic range of
While only strain was directly measured, applied stress was calculated using the Young’s modulus of the epoxy-powder patch. As the patch is a composite sample, Young’s modulus was calculated using the theory of rigid inclusions in a non-rigid matrix. The elastic modulus of particulate reinforced composites can be described using the series and parallel models for the arrangement of phases [18]. A uniform stress/strain field is not sufficient to describe a composite system that has a very soft matrix, with different shapes and arrangements of powders. The model by Guth was based on Einstein’s equation for the viscosity of a suspension of rigid inclusions, introducing a particle interaction term and shape factor for non-spherical powders embedded in soft matrix, like an elastomer [19]. For non-spherical particles the equation is expressed as the following:

$$E_C = E_m \left( 1 + 0.67pV_p + 1.62p^2V_p^2 \right)$$

The shape factor ($p$) was defined as the ratio of particle length to thickness, and $V_p$ is volume fraction of particulate inclusions.

Figure 7 shows magnetic response under tensile stress and strain for the composite samples listed in Table 1. All samples were tested three times and averaged. The B-H plots in Figure 7 were obtained at zero stress, showing a linear response in magnetic flux density as a function of applied magnetic field up to 1500 Oe. The entire measured region is linear because the applied field strength is below the magnetic saturation level. Therefore, the slope corresponds to the low-field permeability at that measured stress. Further increments of tensile stress were applied to the composite patch to get a series of B-H curves at each stress level. The permeability change as a function of applied stress was calculated from the B-H measurements from each applied stress (not shown in Figure 7). The general change of permeability was $\sim$0.3% over the range of applied stresses as shown in the right side of the figure. The sample ‘C1’, which has the softest matrix (low elastic modulus ($E_E$)) shows a negative response in permeability with increasing stress. This is in contrast with the other samples with a stiffer matrix which show increased permeability as the stress increases.

While the negative response was not expected, the soft matrix provides very small strain transfer to the particles. Instead, it just allows for the particle spacing to increase with increasing strain, further decoupling the particles. This decoupling may cause the particles to perform more as individual units rather than one unified set, which will change the shape anisotropy of the system from a disc shape to a sphere. This will correspondingly decrease the permeability, compared to the unstressed state, as observed in Figure 7a. The stress/strain dependence of permeability change ($\Delta\mu$) in Figure 8 was replotted from the permeability plots of Figure 7 and shows a maximum in permeability between 300 and 500 ppm, except for C1. This positive correlation between permeability and tensile strain/stress indicates an enhancement in magneto-elastic coupling in the composite. This coupling in the composite indicates the potential for its use as a magneto-elastic damage detection sensor, measuring the increased stress due to incurred damage. From these initial results, static sensor characteristics can be estimated, although they are not optimized in any way for application as a sensor. Stress sensitivity is highly dependent upon the type of epoxy being used, showing a maximum of $\sim$15.6 GPa/(G/Oe) for composite C4, with values of $\sim$9.2 GPa/(G/Oe) for C3 and $\sim$0.5 GPa/(G/Oe) for C2. All three show a linear trend after a critical applied stress: 0.1 MPa for C2, and $\sim$3 MPa for C3 and C4. The accuracy and precision of these sensitivity values, however, are limited by variation in the strain gages used during the measurement; the strain gages used for this evaluation exhibit ±0.5% variation. Further work will be carried out to extend the applied strain/stress to the plastic region of the aluminum substrate and even to crack initiation and failure, which will be essential for structural health monitoring applications.
accuracy and precision of these sensitivity values, however, are limited by variation in the strain gages used during the measurement; the strain gages used for this evaluation exhibit ±0.5% variation. Further work will be carried out to extend the applied strain/stress to the plastic region of the aluminum substrate and even to crack initiation and failure, which will be essential for structural health monitoring applications.

Figure 7. Magnetic and magneto-elastic interactions of the flake composite samples (C1–C4) listed in Table 1. (a–d) Magnetic flux density and permeability measured as a function of applied tensile stress using a flux meter for integrating the signal from the pickup coil.
3.2. Dynamic Magneto-Elastic Sensing Properties

In addition to the quasi-static measurement of magnetic properties under applied strain, dynamic tests were performed using the ultrasonic guided wave evaluation technique using a PZT actuator and an MPC patch sensor. The PZT actuator was excited at ultrasonic frequencies (e.g., 60 and 120 kHz) to generate a GLW from the center of the plate. The associated guided wave propagation was successfully captured by the MPC patch sensor using the magnetic circuit device as shown in Figure 4. In our previous work, the influence of particle size and shape of Galfenol MPC patches was investigated to optimize directional sensitivity for GLW applications [20]. The MPC patch sensor using Galfenol flakes showed the best sensing performance, exhibiting notable magnetization and magnetostriction even at low field strengths [13,20].

In order to compare the sensing performance between Galfenol and Alfenol alloys, three powder-epoxy composite samples (i.e., MPC patches) were prepared for each alloy, as described above. Figure 9a shows the received GLW signals for the excitation frequencies of 60 and 120 kHz, acquired by the MPC patches. The first noticeable waveform with the highest amplitude for both frequencies was identified as the incident GLW mode arriving directly from the PZT actuation source to the MPC patch sensor. Other waveforms in the signal data are the GLW mode reflections from the circular boundary of the plate structure. The average signal amplitude of the Galfenol MPC patch was a little larger than that for Alfenol, as seen in Figure 9b, exhibiting a 35% and 9% larger signal amplitude at the frequencies of 60 and 120 kHz, respectively. Especially at the higher frequency,
dynamic sensing performance is not significantly different between Galfenol and Alfenol MPC patches utilizing flake-type powders.

Figure 9. (a) Ultrasonic guided wave signal sensing performance at frequencies of 60 and 120 kHz, respectively, for Galfenol and Alfenol particle epoxy composites with a powder weight fraction of 50% and particle sizes of 106–150 μm and (b) peak-to-peak and average values in voltage amplitude for three composite samples of Galfenol and Alfenol.

4. Conclusions

Polymer-bonded composites of an Fe$_{81}$Al$_{19}$ alloy (Alfenol) were fabricated and their magnetic and magneto-elastic sensing performance was investigated as functions of Alfenol powder fraction and shape under applied tensile and compressive stresses. Flake-shaped particles are more sensitive to increases in applied stress than spherical particles, exhibiting a more pronounced permeability increase with an increase in stress. The softest polymer matrix exhibited a negative change in magnetic permeability, possibly due to the decoupling of individual particles and corresponding changes in the shape anisotropy of the system. Epoxies with stiffnesses higher than ~0.1 GPa in elastic modulus showed enhanced magneto-elastic coupling with applied tensile stress/strain. In addition to the static state, Alfenol MPCs with flake-type powders exhibited good dynamic sensing performances similar to Galfenol composites at frequencies of 60 and 120 kHz in ultrasonic guided wave applications.

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