Novel Coiling Behavior in Magnet-Polymer Composites

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Magnet-polymer (Magpol) composites have an interesting ability to undergo large strains in response to an external magnetic field. The contraction behavior of composites of silicone and micron sized iron particles induced by an external magnetic field was studied. Simply by changing boundary conditions, Magpol can exhibit shape change from axial contraction to a novel coiling mechanism due to buckling. The analytically predicted magnetic fields for buckling agreed well with the measured values. The strain versus magnetic field relationship suggested that postbuckling behavior is a stable symmetric bifurcation, which is useful for continuous actuation. The coiling mode of Magpol exhibited high actuation strain (up to 60%) and actuation stress (up to 184 kPa), thus shows good potential for use in soft actuators.

Introduction

A flexible composite of polymer matrix and a magnetic filler exhibits useful and unique transduction properties when placed in an external magnetic field. This type of magnetic-polymer composite (Magpol), also termed ferrogel,[1–8] magnetic gels,[9,10] magnetic field sensitive gel,[11–16] magnetorheological elastomer,[17,18] or magnetoactive polymer,[19] can readily change its shape and mechanical properties upon exposure to an external magnetic field. Many applications have been identified for Magpol including controllable dampers,[20] stiffness tunable supports,[21] miniature fluid pumps,[22] drug delivery,[23] cancer treatment,[23] sensors,[24,25] soft transducers, and artificial muscles.[26]

Actuation occurs because an external magnetic field exerts a force on the magnetic filler particles, causing the movement of the entire composite. This movement of the composite is due to the strong interfacial adhesion between the magnetic particles and the polymer chains in the matrix.[12] Reported modes of shape change in Magpol include deflection,[27] elongation,[14,28] and contraction.[29] Shape change behavior of Magpol can be used for soft actuation, the behavior can be tuned by geometrical and material parameters to exhibit mechanical transition analogous to first or second order phase transformation, hence Magpol can be used for both linear and switching (i.e., on–off) applications.[30]

Previous work has shown that strains of up to 40%[14] and stress of up to 6.8 kPa[28] could be achieved in elongation mode for a composite of poly(vinyl alcohol) gel and Fe3O4 particles. In contraction mode, Magpol was modeled to mimic muscular actuation and shown to produce work of up to 7 mJ.[31]

In this work, the actuation behavior of Magpol in a compressive magnetic field was studied. Different actuation modes were achieved simply by changing the position of the sample with respect to the magnetic field or by controlling the sample deformation in the radial direction (Figure 1). A novel coiling mechanism was observed,
buckling of Magpol in an external magnetic field in a suitably constrained region causes coiling. This is analogous to the mechanically induced helical buckling under compression of a long homogeneous rod in a circular cylinder. A comparison was made between buckling in Magpol and magneto-elastic buckling, the characteristic snapping action of most magneto-elastic buckling phenomena was not observed, instead a gradual deformation was found. Our experimental results showed that coiling possesses higher strain and stress compared to simple axial contraction.

**Experimental Part**

**Synthesis of Magpol Composites**

Spherical iron particles (Alfa Aesar, average size 3 μm) were chosen as the filler material because of their good magnetic properties and ready availability in powder form. Polysiloxane (i.e., silicone) (ECOFLEX-0010, Smooth-On, USA) was selected as the polymer matrix due to its good flexibility and reasonable environmental stability.

In a typical sample preparation, the polymer matrix was prepared firstly by mixing and stirring 10 mL of the precursor and 10 mL of Pt-based curing agent for 5 min. Both the precursor and the curing agent were provided as a package by the manufacturer. Iron powder with weights ranging from 2.3 to 102 g was then added to the polymer compounds in steps of 1 g while mixing was carried out by hand. The process typically took 15–30 min depending on the intended filler concentration. The mixtures were then subjected to vacuum to remove residual bubbles, and transferred into a cylindrical mold ($d = 11$ mm and $L = 130$ mm). The samples reached their final shape and mechanical properties after curing for 24 h at room temperature.

**Characterization**

The cross-section of the samples was prepared by cutting the samples by a sharp pen knife. The morphology of the composites was then studied using a 6340F scanning electron microscope (SEM) without surface coating. Particle size was analyzed using ImageJ software. The magnetic properties of the particles and the composites were determined by a Lakeshore 7404 vibrating sample magnetometer (VSM). The room temperature magnetization curves were fitted by a mathematical function using TableCurve2D software. The mechanical properties of the composite were tested according to ASTM D 638-03 standard using an Instron 5567 tensile tester.

**Experimental Setup**

The schematic of the experimental setup to test the actuation properties of Magpol is shown in Figure 2. The magnetic field was generated by a Lakeshore CM-4 dipolar electromagnet, the magnetic field strength can be varied by altering the current passing through the electromagnet coils. One end of the sample was fixed by an aluminum holder while the other end was free. The displacement of the sample was guided by a cylindrical glass tube, both the sample surface and the glass tube were lubricated by a low viscosity machine oil ($n = 9.6 \text{ m}^2/\text{s}$) to minimize friction. The sample displacement was recorded by an Acuity AR600 triangulation laser displacement sensor with resolution of 61 μm. The force generated was recorded by a Vernier dual range force sensor (resolution of 0.01 N) attached to the free end of the sample.

**Finite-Element Simulation**

Magneto-structural finite element simulation was performed using ANSYS Multiphysics version 10 to provide qualitative...
understanding of the initial stage of the coiling process. Coupled-field SOLID98 elements were used to model the composite sample, air and the electromagnet. The use of coupled-field elements allowed simultaneous solving of magnetic and structural models when spatial displacement (UX, UY, UZ) and scalar magnetic potential (MAG) degrees of freedom were activated. The experimental values of sample dimensions were used. The two magnet coils of the electromagnet were modeled by two racetrack coils of the same dimensions. A spherical air region with diameter 60 times the size of the air-gap was modeled to sufficiently cover the entire model and simulate far-field condition. The mesh size was refined at the sharp edges of the models, and especially in the air-gap region. The magnetization curve [Figure 3(b)] and the Hookean elasticity of the composite sample (from Figure 4) determined experimentally were used in the model. The effect of gravity was included as well as the experimental value of sample density. The spatial constraint of the glass cylinder was not modeled since only the initial stage of sample deformation was simulated. Parallel field boundary condition was applied at the outer surface of the air region.

**Results**

**Physical Properties of Magpol**

Scanning electron micrographs of cross-section of silicone-iron Magpol in both secondary electron and backscattered electron modes are shown in Figure 5. Iron particle aggregates with average size ranging from 0.1 to 5 μm were observed, the aggregate distribution was reasonably uniform due to thorough mixing during sample preparation.

Room temperature magnetization curves of (a) the iron filler particles and (b) silicone–50 wt.-% (12 vol.-%) Fe composite. The inset shows the magnetization saturation of the composites at different Fe concentrations.

![Figure 3. Magnetization curves of (a) the Fe particles used and (b) Silicone–50 wt.-% (12 vol.-%) Fe composite. The inset shows the magnetization saturation of the composites at different Fe concentrations.](image)

![Figure 4. Tensile properties of Magpol of various filler concentrations.](image)

![Figure 5. Scanning electron micrographs of a silicone–20 wt.-% (3 vol.-%) Fe sample. (a) Secondary electron micrograph and (b) backscattered electron micrograph.](image)
samples are shown in Figure 3. The iron particles exhibited typical soft ferromagnetic properties characterized by a nonlinear magnetization curve with a small hysteresis. The measured value of susceptibility of the iron particles was 18, consistent with previously reported values.\textsuperscript{[41,42]} The silicone–50 wt.-%(12 vol.-%)Fe samples exhibited qualitatively similar magnetic properties to those of the iron particles. The saturation magnetization ($M_S$) value of the composite increased linearly with the concentration of the magnetic particles, suggesting that particle aggregation did not significantly affect the magnetic properties of the composites. The magnetization curves $M(B)$ of Magpol samples were satisfactorily fitted by a sixth order polynomial function.

The tensile properties of the pure silicone as well as Magpol samples containing different filler concentrations of iron particles are shown in Figure 4. The addition of particles increased the modulus of the composite, the range of elastic behavior decreased with increasing particle concentration. However, composites at all particle concentrations studied in this work remained elastic up to a tensile strain of 100%.

Coiling Phenomenon Observed in Magpol

In the experimental setup, a cylindrical glass tube was used to guide the sample and to prevent it from sticking to the magnet poles. Interestingly, changing the diameter of the tube, $d_{\text{tube}}$, resulted in different modes of shape change. When the ratio of tube diameter to sample diameter, $k = d_{\text{tube}}/d_{\text{sample}}$, was set at 1.2, axial contraction was observed [Figure 1(b)]. For a large glass tube of $k = 1.6$, coiling was observed [Figure 1(c)]. A hybrid mode [Figure 1(d)] was observed when $k = 1.4$. The different stages of coiling action are shown in Figure 6. During the first stage, the sample contracted slightly under the magnetic field [Figure 6(a)]. Beyond a critical value of magnetic field, the samples buckled [Figure 6(d)], further increase in the magnetic field led to coiling of sample to conform to the space within the tube [Figure 6(g) and (h)].

Magneto-structural finite element simulation was performed using ANSYS. The results [shown in Figure 6(b), (c), (e), and (f)] revealed that coiling of Magpol is caused by local buckling in the high magnetic field intensity region between the magnet poles. In this region, the magnetic field gradient is largest and thus the force is greatest. The compressive magnetic force in this region caused the flexible composite structure to buckle.

Characteristics of Coiling Actuation of Magpol

The shape deformation of Magpol induced by an external magnetic field can be used for useful actuation. The response time of the samples to the change in magnetic field is less than 60 ms, in agreement with a previous study which reported a response time under 100 ms.\textsuperscript{[14]} The dynamic properties of Magpol in elongation mode were studied using a different electromagnet, deformation of more than 24 000 cycles at 80 Hz can be achieved without a significant shift in strain-field characteristics. In general, it is known that field driven actuators like Magpol have fast switching time and better service life than ionic actuators.\textsuperscript{[43]} The coiling strain, defined as the change in z coordinate of the free end divided by the initial sample length, is plotted as a function of the magnetic field for silicone–60 wt.-%(17 vol.-%)Fe samples [Figure 7(a)]. The plot is smooth and characterized by three stages. The first stage involved a small decrease in sample length, the second stage started with sample buckling, which led to coiling in the region between the coils. The coil then propagated from the region between the electromagnet poles toward the free end of the sample. The last stage was the reduction in coil height due to the attraction to the region of high magnetic field gradient (i.e., the region between the poles), this process is similar to the compression of a spring. This process continued until each ring of the coil physically touched each other.

As the magnetic field was decreased, the coil structure relaxed and eventually uncoiled to return to the original straight shape. This actuation exhibited hysteresis, that is, the shortening and relaxing strain values were not equal at the same field strength. The strain versus magnetic field for axial contraction of the same samples is shown in Figure 7(b). For a given magnetic field strength, the axial contraction strain was lower than the coiling strain. The maximum coiling and contraction strain as a function of filler concentrations are shown in Figure 7(c). The coiling strain was higher than contraction strain by approximately 50% for all concentrations.

The actuation stresses produced by silicone–83 wt.-%(40 vol.-%)Fe samples in axial contraction and coiling modes are represented by surface graphs in Figure 8. The relationship of the actuation stress with the change in length ($\Delta L$) and the initial sample length ($L$) was studied. The results revealed that the actuation stress exhibited similar behavior in both coiling and axial contraction modes. The stress values decreased as $\Delta L$ increased. This is similar to the length-tension behavior found in natural muscles.\textsuperscript{[44]} The relationship of the maximum value of actuation stress with the initial sample length exhibited a peak-like behavior. This suggested that there is an optimal sample length which yields highest actuation stress. Here the medium sample length corresponding to the highest maximum stresses (184 kPa for coiling and 164 for axial contraction) was referred to as the optimal length.
Figure 6. The coiling behavior of silicone–20 wt.-% (3 vol.-%) Fe samples at different stages as observed by experiment and simulation: (a) \(B = 0.11\) T, the sample remained straight, stress builds up in the sample; (b) and (c) Magnetic field density (\(\text{A} \cdot \text{m}^{-1}\)) plot and stress (Pa) distribution plot obtained from FEM simulation when \(B = 0.11\) T; (d) \(B = 0.216\) T, the sample started to bulge; (e) and (f) Magnetic field density plot and stress distribution plot obtained from FEM simulation when \(B = 0.216\) T; (g) \(B = 0.324\) T and (h) \(B = 0.433\) T, The sample coiled due to the cylindrical spatial constraint; (i) \(B = 1\) T. The coiling configuration reached its limit.
Discussions

Physical Properties of Magpol

Important properties relevant to the use of Magpol for actuator applications include mechanical and magnetic properties. Mechanical properties of interest include high flexibility, strain and load carrying capacity; magnetic properties of interest include high magnetic susceptibility ($\chi$) and $M_S$. These properties, in turn, depend on the polymer matrix and the type and concentration of the magnetic filler. Based on these requirements, silicone elastomer and iron particles were chosen as the constituent components for Magpol. Regarding the choice of particle size, as the size of the particles gets smaller, interesting properties can be observed such as superparamagnetism but the value of $M_S$ usually decreases significantly due to spin disorder at the particle surfaces. Small size of the filler particles will also lead to aggregation and can reduce interfacial adhesion.

![Figure 7.](image1)

**Figure 7.** The actuation properties of Magpol: (a) Coiling strain of silicone–60 wt.-%(17 vol.-%)Fe samples as a function of magnetic field; (b) axial contraction strain of silicone–60 wt.-%(17 vol.-%)Fe samples as a function of magnetic field; (c) maximum coiling and contraction strains as a function of filler concentration.

![Figure 8.](image2)

**Figure 8.** Isometric axial contraction stress and coiling stresses as a function of change in length and sample length of silicone–83 wt.-%(40 vol.-%)Fe samples, represented by surface graph and contour graphs. (a) Coiling stress and (b) axial contraction stress.
between particles and the polymer matrix, thus undesirable particle migration within the matrix may be caused by strong magnetic forces.\(^{[5]}\) On the other hand, as the relative size of the particles and the sample increases, it is difficult to achieve homogeneous distribution of particles. Therefore, the iron particles were chosen in the micron size range.

Agglomeration does not significantly affect the magnetic and mechanical properties. The composites show a similar shape of magnetization curve to that of iron particles and their \(M_S\) values increase linearly with filler concentration. The composites exhibited elastic behavior up to 100% strain over all filler concentrations studied in this work.

**Coiling Phenomenon of Magpol**

The sample contracted and buckled under the compressive magnetic force prior to the coiling shape change. Subsequent increase in the magnetic field made the sample coil within the glass tube. Continuous application of force on a rod-like object after buckling within a cylindrical constraint has been shown to lead to a helical shape.\(^{[46]}\) Due to the high flexibility and strong response to magnetic field, coiling is the natural contraction mechanism for Magpol. However, when the diameter of the cylindrical constraint is small, pure axial contraction instead of coiling was observed due to the lack of radial displacement. Therefore, two criteria are needed for coiling. The first is that the composite undergo buckling, the second is the appropriate diameter of the cylindrical constraint. The following section provides a basic framework for the analysis of magneto-elastic buckling of Magpol composites.

**Buckling Analysis of Magpol**

Buckling of Magpol composites can be analyzed by first considering the load acting on the samples, this load is the balance of the magnetic force and the sample weight (Figure 2):

\[
P = F_{\text{magnetic}} - W_{\text{weight}}
\]

where \(F_{\text{magnetic}}\) is the magnetic force acting on Magpol sample and can be expressed as:\(^{[47]}\)

\[
F_{\text{magnetic}} = \int \nabla M \cdot B \, dV = \int_{z_{\text{bottom}}}^{z_{\text{top}}} \frac{\partial B}{\partial z} \, a \int M(B) \, dB
\]

where \(a\) is the sample cross-sectional area. The weight of the sample, \(W_{\text{weight}}\), and can be computed as:

\[
W_{\text{weight}} = \int g \, dm = \int \rho \, dVg = \int_{z_{\text{bottom}}}^{z_{\text{top}}} \rho \, gdz
\]

As buckling occurs at small strain, the cross-sectional area can be assumed constant. Therefore, the load can be expressed as:

\[
P = a \int_{B_{\text{bottom}}}^{B_{\text{top}}} M(B) \, dB - a \int_{z_{\text{bottom}}}^{z_{\text{top}}} \rho \, gdz
\]

The \(M(B)\) function can be fitted by a sixth order polynomial function and the magnetic force can be computed from Equation (2). The magnetic field \(B_{\text{bottom}}\) (at \(z_{\text{bottom}}\)) and \(B_{\text{top}}\) (at \(z_{\text{top}}\)) were experimentally measured and thus the load \(P\) can be calculated.

Euler’s formula, which states that buckling of a long sample occurs when the load acting on it is equal or more than a critical value \(P_{\text{cr}}\), was applied. \(P_{\text{cr}}\) can be expressed as:\(^{[48]}\)

\[
P_{\text{cr}} = \frac{\pi^2 EI}{(KL)^2}
\]

where \(E\) is the Young’s modulus of the composite, \(I\) is the area moment of inertia of the sample, \(L\) is the unsupported length of the sample, and \(K\) is the effective length factor.

The samples had one end fixed and is loosely constrained in a cylindrical tube, hence there are two relevant cases for buckling analysis. One is fixed and free end model (\(K = 2\)) and the other is fixed and pinned end model (\(K \approx 0.7\)).

From Equation (1) and (5), it can be seen that buckling occurs when the magnetic force exceeds the sum of the sample weight \(W_{\text{weight}}\) and the load \(P_{\text{cr}}\). From this result, the critical magnetic force can be calculated and the value of the critical buckling magnetic field \(B_{\text{cr}}\) can be predicted. Figure 9 shows that the predicted magnetic field for buckling of Magpol, for example, silicone–20 wt.-%(3 vol.-%)Fe samples, agrees well with the experimental \(B_{\text{cr}}\) value.

The load \(P\) was calculated from Equation (1) and (5) using the experimental magnetic field values. This load was compared to the theoretical critical load values for the following two scenarios of buckling. One is full sample buckling (effective buckling length = sample length) and the other is local buckling near the magnet pole region (effective buckling length = magnet pole diameter) (Figure 10). It can be seen that the buckling should occur in the second case because the experimental load values lies in the range limited by the predicted load curves. This is in agreement with experimental observations and is supported by the magneto-structural simulation.
Comparison of Buckling in Magpol with Other Magneto-Elastic Buckling Phenomena

The locally curved buckling shape is similar to kink instability found in flexible current carrying conductors under self-generated fields\(^{35}\) or the instability of a helical wire carrying current in a longitudinal magnetic field\(^{36,37}\). The overall coiling shape is similar to that of a ferromagnetic whisker after elastic buckling under compressive force in a longitudinal magnetic field\(^{37,38}\).

The continuous change in length shown in Figure 7(a) suggests that Magpol has a stable postbuckling behavior which can be characterized by a stable symmetric bifurcation\(^{49}\). This behavior is similar to the buckling of column structures. In contrast, many other types of magneto-elastic buckling exhibit unstable postbuckling characterized by a limit point beyond which an abrupt change in shape or "snapping" action takes place\(^{39,40}\). The smooth coiling shape change makes Magpol suitable for actuator applications. Another advantage of symmetric bifurcation is that the system is relatively insensitive to material inhomogeneity or misalignment\(^{39}\).

Coiling Magpol as an Actuator

Among the three stages of coiling process of Magpol, the second stage which involves the formation of the coil is the most important stage as it contributes more than two-thirds of the overall strain. This stage can be achieved by a magnetic field strength of about 0.5 T which is easily found in common permanent magnets. The actuation strain and stress produced by Magpol in coiling mode are higher than those achieved in simple axial contraction mode and previously reported values\(^{28,50}\). It is noteworthy that the coiling strain and stress are also higher than the maximum strain and stress generated by skeletal muscles\(^{26}\). This considerable improvement in actuation performance can enable a higher feasibility and allow a wider range of applications of actuators based on Magpol.

Conclusion

Composites of silicone elastomer and iron particles were synthesized, characterized, and their shape changing behavior in an external magnetic field was studied, a coiling mode of shape change was found and investigated. Large strain (up to 60%) and stress (up to 184 kPa), fast response time (less than 100 ms) observed in this mode of shape change show that actuators based on this class of materials can be used in many technological applications. Also, the low Young’s modulus values, typically less than 0.06 MPa, shows promising potential for soft actuators or artificial muscle applications. Further research will focus on lowering the magnetic field requirements for a more efficient control of this system.
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