WORK LOOP AND ASHBY CHARTS OF ACTIVE MATERIALS

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FINAL TECHNICAL REPORT

Executive Summary

The use of active materials for structural applications is of interest in the context of structural health monitoring and the use of adaptive/morphing/shape changing structures for aerospace structures in UAVs. Magnet-Polymer composite materials (Magpol) can be used for such applications. One of the areas that is of considerable interest is how much work can be done by Magpol and where the performance metrics of Magpol (e.g., actuation stress, actuation strain) place this class materials in the relevant Ashby charts. Hence the motivation of this work is to develop a novel work loop method for determining the actuation performance of Magpol and to construct Ashby charts.

Under this grant, we have conducted detailed studies of the work-loop characteristics of light weight active polymer-magnet structural materials and placed the actuation properties of these materials in the relevant Ashby charts.

1. Introduction

Active materials are capable of undergoing a change in their properties under the influence of an external stimulus. This can include a change in shape, mechanical properties, chemical, electrical properties etc. The use of active materials in structural applications usually involves the ability of the material to actuate, sense changes in temperature, pressure, damage etc. and

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14. ABSTRACT

The use of active materials for structural applications is of interest for structural health monitoring and as adaptive/morphing/shape changing materials found in aerospace structures. In this project detailed studies of the work‐loop characteristics of active polymer‐magnet composite materials (Magpol) were conducted and Ashby charts constructed to show performance metrics (e.g., actuation stress, actuation strain, self-healing) of iron-loaded compositions compared to other active materials. Multiple actuation modes, including novel coiling behavior, were observed in the composite systems under study. Magol maximum actuation frequency for dynamic properties of are up to 80 Hz, which is higher than earlier reported values for PVA hydrogel ‐ Fe3O4 composites. In contrast to conducting polymer actuators that show shifting of strain profile and short life (<10,000 cycles for strains of 2%), Magpol composites actuate up to 24,000 cycles at 80 Hz without change in strain characteristics. Self-healing of Magpol prepared using ferrite nanoparticles of different Curie temperatures was examined; after magnetic heating, near complete recovery was observed even after repeated damage.

15. SUBJECT TERMS
functional composites , Active Materials, Magnetic Activity

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undergo some amount of recovery. Actuation, the ability of a material to respond to an alteration in its environment by mechanical means, is a common occurrence in nature and forms the cornerstone of living systems. Actuators can be defined as controllable work-producing devices. Actuators are based on a large variety of physical principles and exhibit a wide array of performance capabilities. There is a current unmet need in actuation technologies, i.e., actuators do not provide soft, smooth motion.

An added advantage to actuator performance would be their ability to “morph”, i.e., to shape change under an appropriate stimulus, such as external electrical, magnetic, chemical or thermal stimuli. Morphing can be described as ‘efficient adaptability that may include micro, macro, structural and/or fluidic approaches. This “intelligence” can be of great use in fields such as biomedicine, structural health monitoring, defense, aerospace, adaptive optics, robotics, etc. Examples include aircrafts with morphing wings to optimize flight conditions, expandable stents for use in medical devices, and artificial muscles.

Magnet filler–polymer matrix composites (Magpol) are a class of materials that can be adapted to develop multifunctional composites, due to the considerable flexibility in the choice of the polymer matrix and type of fillers. Advantages of Magpol based systems include remote contactless magnetic actuation, a variety of actuation modes, high actuation strain, self-sensing as well as quick response. The ability of the composite to actuate in a magnetic field as well as to generate heat in an ac field makes this an ideal system to study multifunctionality [1].

1.1 General introduction

Magnet filler–polymer matrix composites (Magpol) have shown great promise in the development of active materials. Previous research has already demonstrated the feasibility of Magpol as an actuator; reported actuation stress values of Magpol surpass that of mammalian skeletal muscles. Materials with properties and constituents similar to Magpol include ferrogels, magnetic gels, magnetic field sensitive gels, magnetorheological elastomers and magnetoactive polymers. The large range of properties that Magpol is capable of exhibiting is due to its flexible nature with respect to the choice of its component matrix and filler. Different groups have studied hydrogels, silicones, polyurethane and rubber as matrices for the development of Magpol; Similarly, the magnetic filler can be magnetically soft or hard depending on the requirements. Thus the properties of Magpol can be tuned by flexibility in choice of polymer and filler.

Our work in a previous project has shown that Magpol can exhibit several actuation modes. These modes include elongation, contraction, deflection and coiling (Figure 1). By changing the boundary conditions, the actuation mode can be changed from axial contraction to a novel coiling mechanism [2]. The different actuation modes of Magpol can be used to perform work, which is measured via the work-loop method. Different actuation modes will produce different force strain characteristics which influences the final work density that can be obtained [3]. The contraction and coiling modes were used to generate work loops and the work density calculated for different sample lengths and filler loading percentages to obtain the maximum work possible [4]. The dynamic response of the Magpol actuators was also measured.

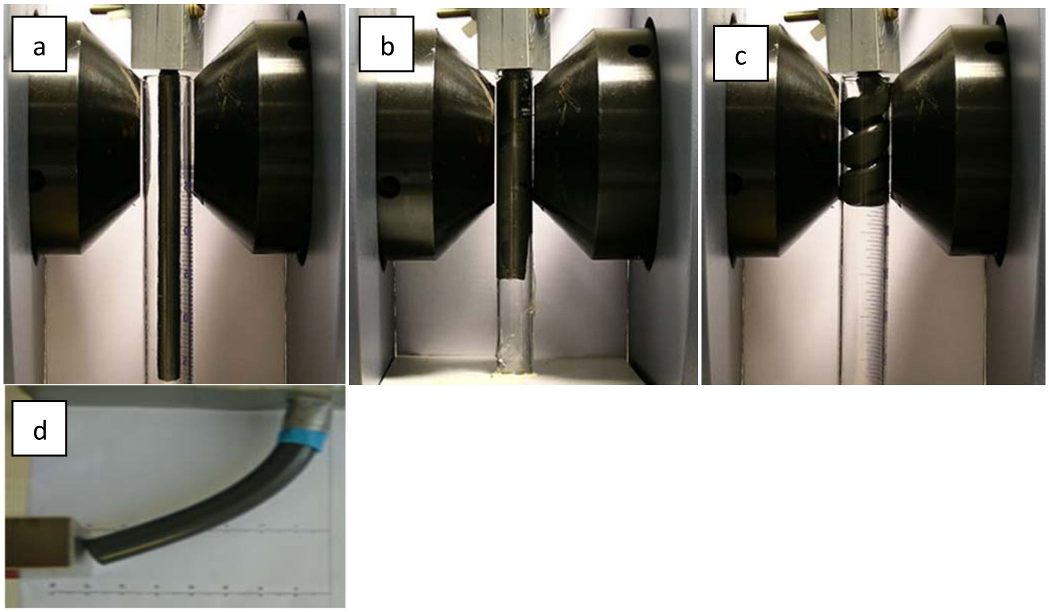


Figure 1: Various actuation modes of Magpol a) Elongation b) contraction c) Coiling d) Deflection[3]

1.2. Novelty

The work loop method is usually used to assess soft biological materials such as muscles and tissues, here it is used for the assessment of inorganic soft materials, i.e., Magpol. The significance of this work is that we have an experimental determination of the stress, strain and work capabilities of Magpol as well as a facile and ready method of placing this material in the context of other actuators such as piezoelectric actuators, shape memory actuators etc.

2. Experimental procedures

2.1. Synthesis of magnet-polymer composites

Spherical iron particles (ave. size 3 µm) were chosen as the filler material, polysiloxane (i.e., silicone) was selected as the polymer matrix due to its good flexibility and reasonable environmental stability. Self healing Magpol was synthesized by substituting the matrix for thermoplastic polymer, while the filler used was ferrite nanoparticles and samples were synthesized via solution casting method.

2.2. Experimental setup to measure actuation

The magnetic field was generated by a Lakeshore CM-4 dipolar electromagnet, the magnetic field strength can be controlled by varying the current through the electromagnet coils. One end of the sample was fixed to an aluminum holder while the other end was free. The sample was placed within a cylindrical glass tube, the sample surfaces and the glass tube were lubricated by a low viscosity machine oil ($\nu = 9.6$ m2/s) to minimize friction. Different actuation modes were obtained by setting appropriate sample positions; the sample elongated when the free end was above the middle of the pole pieces and contracted or coils when the free end was below the middle of the electromagnet. The sample displacement was measured using an Acuity AR600 triangulation laser displacement sensor (resolution of 61 µm) and the generated force was recorded using a Vernier dual range force sensor (resolution of 0.01 N). The force sensor was attached to the fixed end of the sample in elongation mode and to the free end in contraction and coiling modes

2.3. Work loop measurements

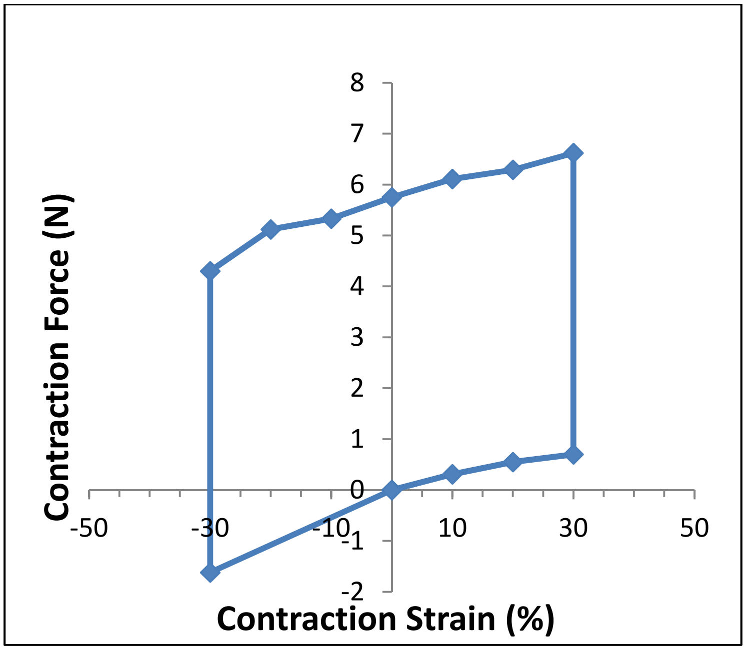
The work done in actuation modes were studied using the work-loop method [Josephson, R.K., J. Exptl. Biology, 1985. 114(1): p. 493]. The samples were subjected to repeated cycles of lengthening and shortening and the force will be measured. The actuation strain was set at 70% of the maximum strain for each mode, e.g, 30% strain in contraction mode and 40% strain in coiling mode, respectively. A magnetic field of 1.5 Tesla was applied (removed) at the end of the lengthening (shortening) phases. Sample work loops were measured at a frequency of 0.5 Hz since work capacity was the measurement of interest. In work loop experiments, net work per cycle usually decreases

2.4. Dynamic properties measurement

For testing of dynamic actuation of Magpol, a similar experimental setup was used. However, the magnetic field was generated by a air-cooled Phywe 06480.01 dipolar electromagnet driven by a Dynatronix DPR40-30-100 pulse power supply, the magnetic field strength and frequency were varied by changing the voltage pulses generated by the power supply. The current was measured by a Fluke 43B power analyzer, the magnetic field strength was determined by a calibrated field - current curve.

3. Results

Figure 2 shows the typical work loop of Magpol in contraction and coiling actuation for silicone - 50 wt% Fe samples while the area of the loop yields the work produced. Table 1 shows the work and work density (work per unit mass) of silicone - 50 wt% Fe samples of various sample lengths. It was observed that for contraction and coiling stress, there is an optimal sample length for which the highest work density value was obtained. For contraction, the optimal sample length was observed to be *L/Lmax* = 0.7; while for coiling, *L/Lmax*= 0.8 as a longer sample is required while coiling.



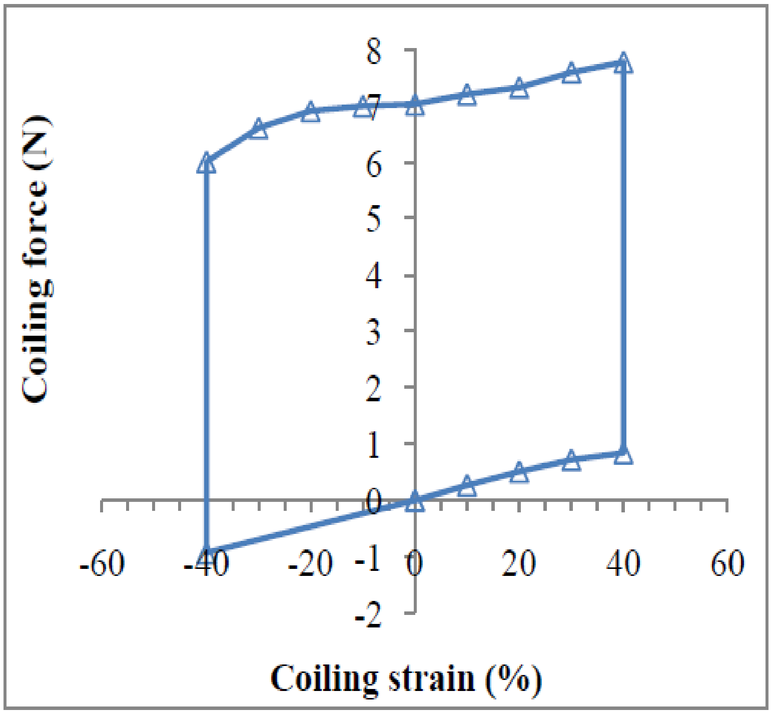


Figure 2 Work loops of silicone - 50 wt% Fe samples with length L/Lmax = 0.8 in (a) contraction and (b) coiling modes[3]

Table 1 Work and work density of silicone - 50 wt% Fe for various sample lengths[3]

Sample length L/Lmax	Contraction		Coiling	
	Work (J)	Work Density (J/kg)	Work (J)	Work Density (J/kg)
1	0.27	11.60	0.52	22.60
0.8	0.35	19.70	0.56	31.42
0.7	0.36	23.65	0.41	27.12
0.5	0.24	19.44	0.29	23.45

Figure 3 shows work density values of silicone - iron Magpol for various filler concentrations in contraction and coiling modes. The work density values increased with filler concentration.

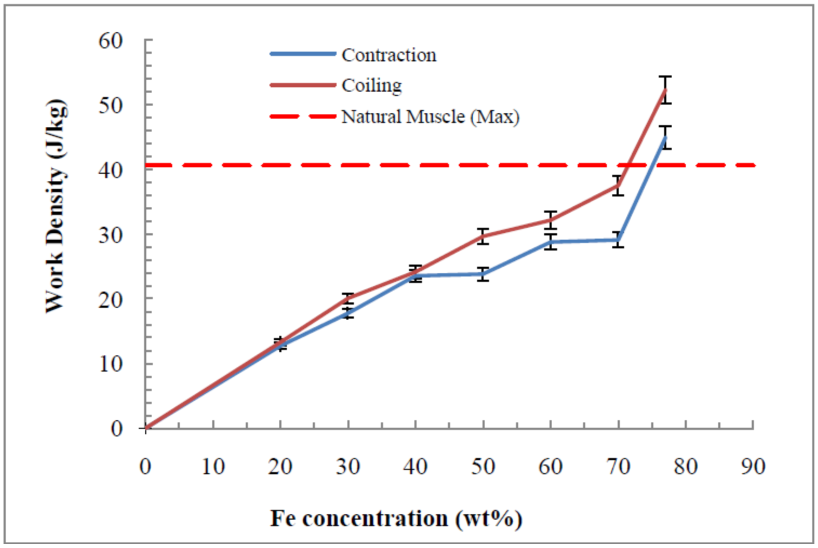


Figure 3 Work density of Magpol with various filler concentration produced in contraction and coiling mode.[3]

The results from this work demonstrated the feasibility of Magpol as a soft actuator. The actuation performance of Magpol showed considerable improvement compared to previously reported values. The strain obtained in the coiling mode was 50% higher than that of contraction. The actuation stress values of Magpol exceed that of mammalian skeletal muscles. It can be seen that the improvements in Magpol performance reported in this work has significantly increased the material's competitiveness, especially among magnetically driven actuators. The actuation strain of Magpol is highest among the members in this group. Magpol has higher actuation stress than solenoids and moving coil actuators and work capacity is 164 kJ/m^3, better than the work capacity of 156 kJ/m^3 achieved by twin boundary reorientation magnetic SMAs.[3]

Two important actuation metrics, force and strain, of Magpol and other common actuators were plotted on an Ashby property chart

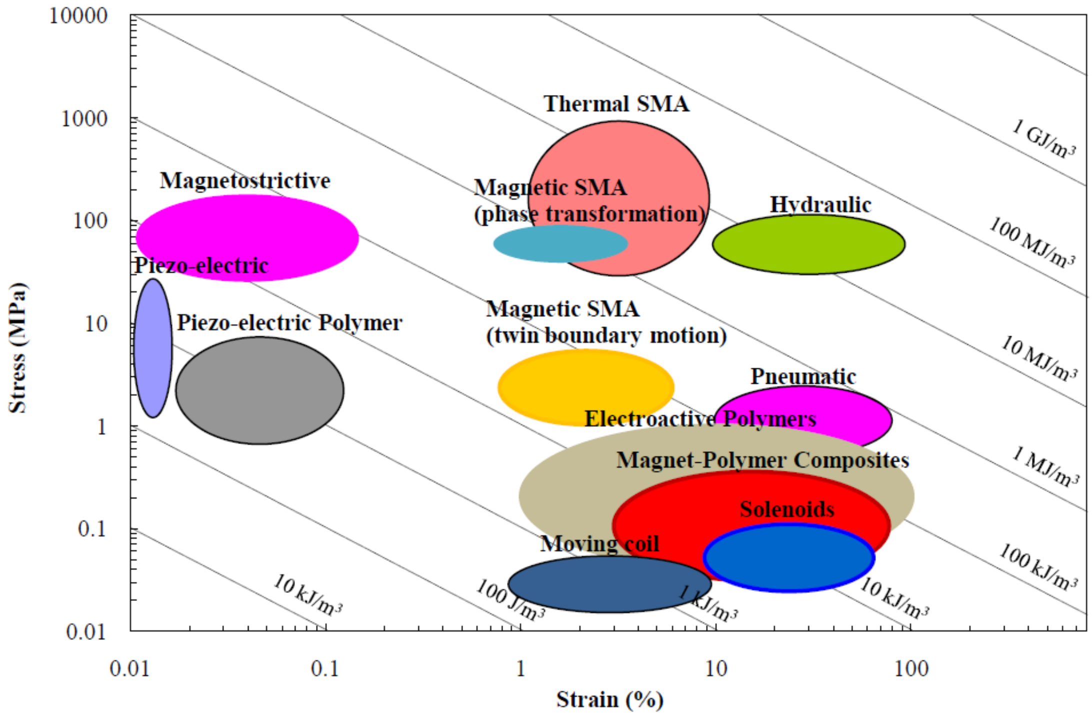


Figure 4 Stress vs. Strain characteristics of Magpol and other actuation technologies[1]

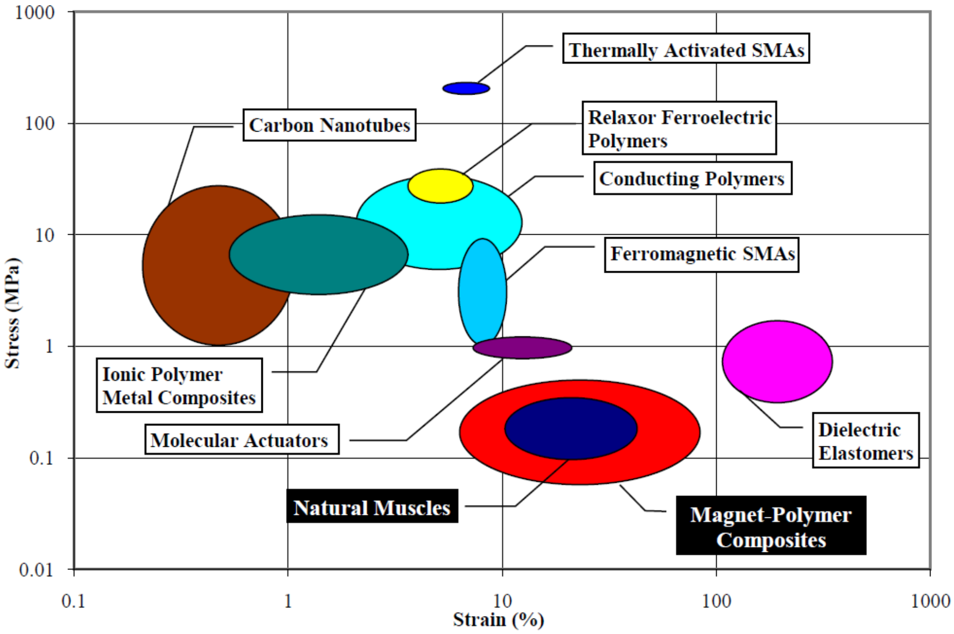


Figure 5 Force vs. Strain characteristics of Magpol and other Polymer Actuators[1]

The work capability of Magpol was also found, for the first time, to exceed that of natural muscles, making this class of actuator attractive for artificial muscles applications. A study of artificial muscles based on carbon nanotube aerogel sheets reported actuation strain of up to 220%, isometric force of up to 3.2 MPa and high frequency of up to 1100 Hz. However, this material exhibited rather limited work capacity (less than 30 J/kg) and a single lateral mode of actuation. Magpol offers an attractive alternative for artificial muscle applications with its low cost, attractive actuation performance (strain, stress and work equal or higher than those of natural muscles) and multiple actuations modes.

The maximum actuation frequency achieved in the study of dynamic properties of Magol is 80 Hz, higher than earlier reported values for a PVA hydrogel - Fe_3O_4 composites. Magpol can actuate up to 24,000 cycles at 80 Hz without change in strain characteristics. This is in contrast to conducting polymer actuators which reported shifting of strain profile and short life (<10,000 cycles for strains of 2%). composition (e.g., type and concentration of magnetic fillers and the type of polymer matrix). The strain rate and strain of Magpol and other competing artificial muscles are shown on a property chart in figure 6, while frequency of operation is compared in figure 7.

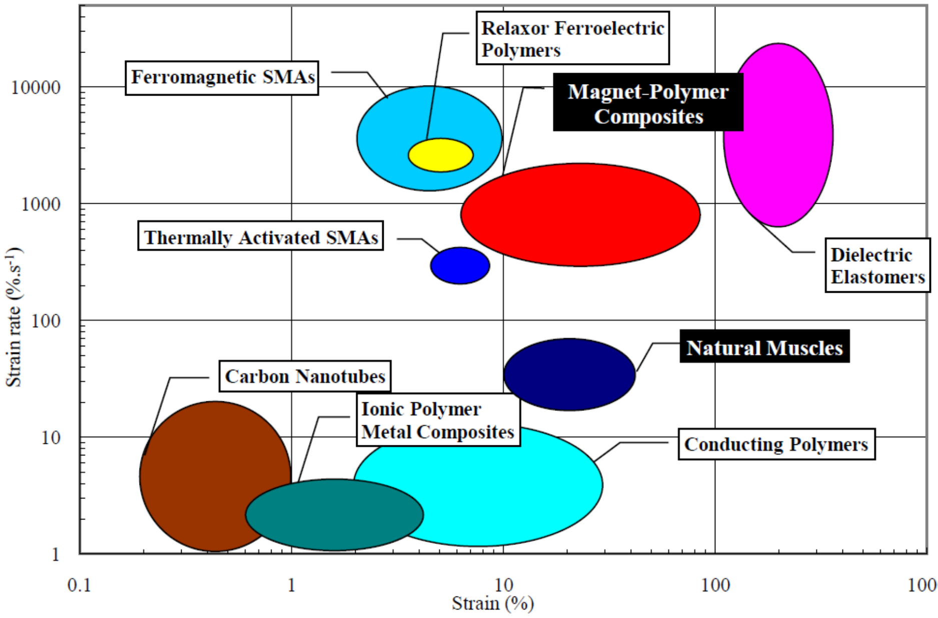


Figure 6 Dynamic actuation characteristics of Magpol and other Polymer Actuators. Strain rate vs. strain[1]

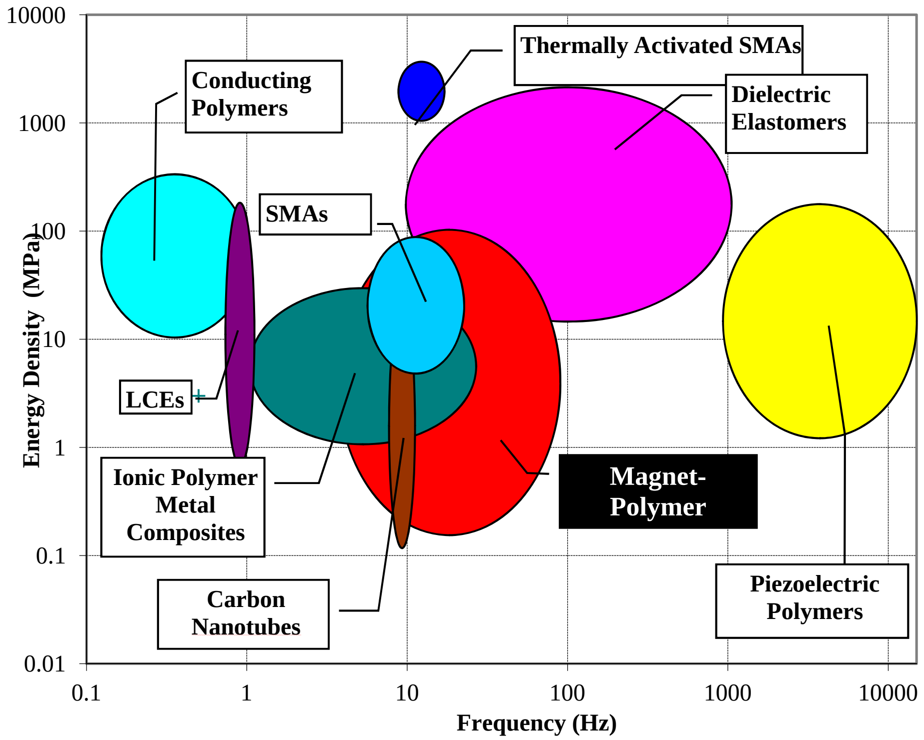


Figure 7 Dynamic actuation characteristics of Magpol and other Polymer Actuators: Frequency vs. Energy density *(unpublished data)*

Another advantage of Magpol actuators is the ability to operate in the absence of an integrated driving source. This extends it application space to areas that require contactless, noninvasive control, such as biomedical implants and aerospace devices.

The idea of using heat to heal thermoplastic polymers can be derived from the fusion bonding or welding processes in thermoplastic polymers, which essentially follow the same principles. The use of Ferrite magnetic nanoparticles embedded in the polymer provides the option of remote heating via the application of a magnetic field. Ferrite nanoparticles with different Curie temperatures were synthesized and their ability to heal tears and cuts in the Magpol film were studied. The healing ability of Magpol as compared to other available polymer self healing systems is compared in Figure 8. Healing efficiency is plotted against time required to achieve maximum healing. The ability of Magpol to undergo repeated damage and healing cycles was also studied and the results are shown in figure 9. It was seen that Magpol was able to achieve almost complete recovery even after repeated damage.

Thus Magpol is an ideal system to incorporate multiple properties to construct a truly active material.

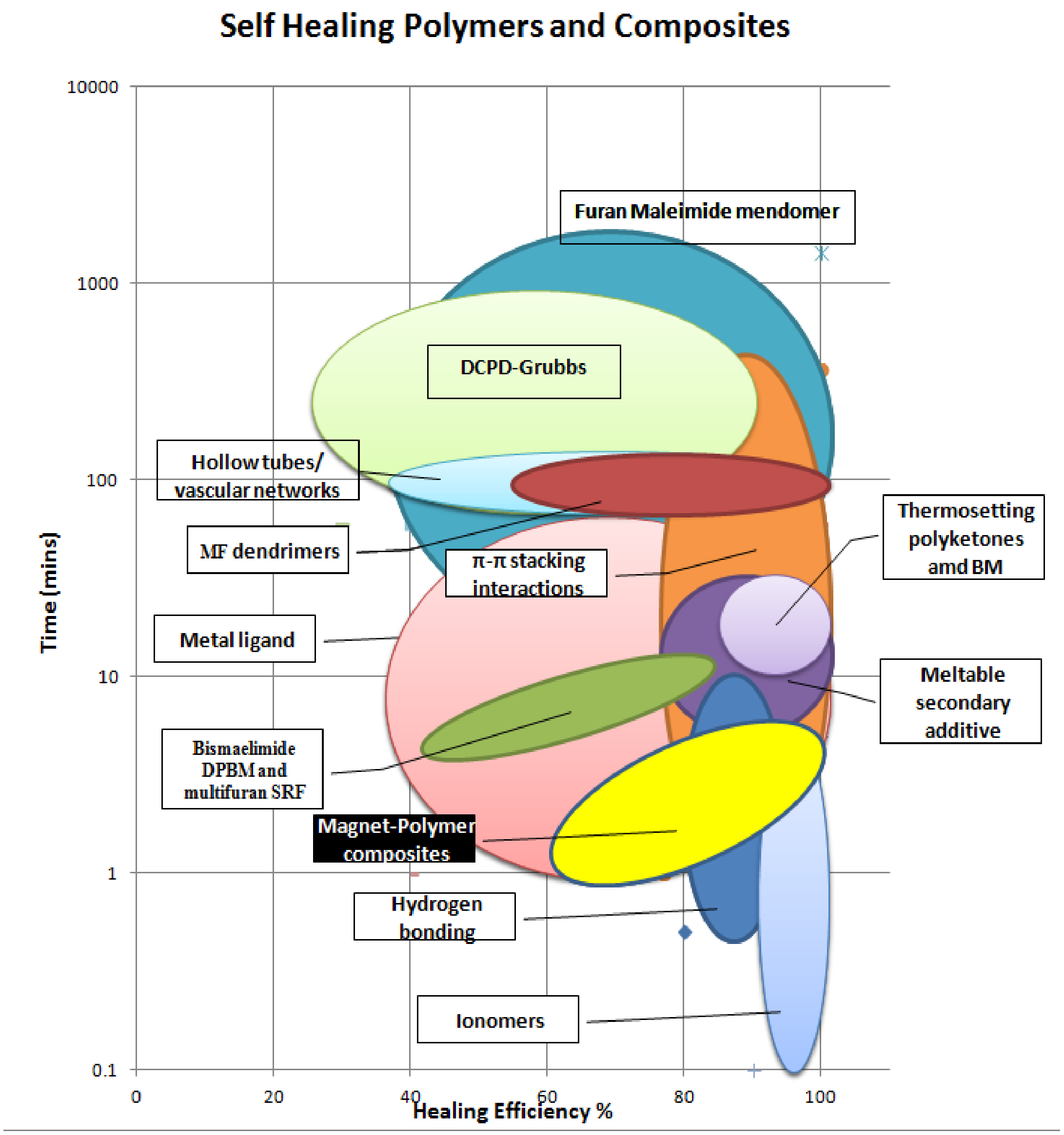


Figure 8: Healing Efficiency vs. time of various self healing material systems *(unpublished data)*

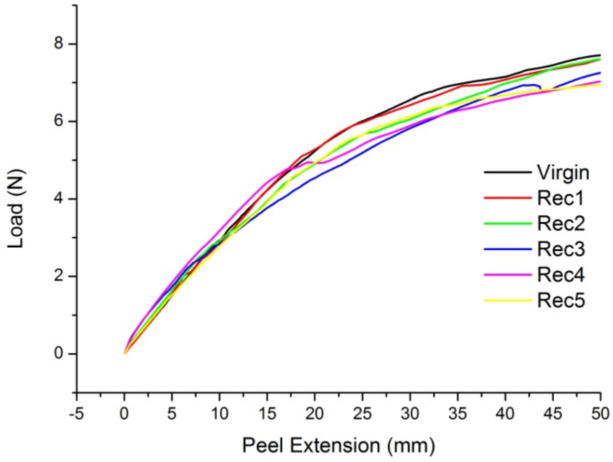


Figure 9 Tear test curves of Magpol for 5 damage and healing cycles *(Unpublished data)*

4. Conclusions

- A Variety of actuation modes, including novel coiling behavior was observed in the Magpol samples
- The maximum strain measured was ~ 80% for elongation. This is the highest among magnetically driven actuators
- The actuation properties of Magpol were compared to other available actuators and also with competing polymer actuator technologies by construction of Ashby charts
- First study of work measurement in Magpol using work-loop method was conducted and it was seen that the work capability measured through work loop was comparable to magnetic SMAs
- Ashby charts were also prepared to compare Magpol with other self healing materials

5. References

1. Nguyen, V.Q., A.S. Ahmed, and R.V. Ramanujan, *Morphing Soft Magnetic Composites.* Advanced Materials, 2012. **24**(30): p. 4041-4054.
2. Nguyen, V.Q. and R.V. Ramanujan, *Novel coiling behavior in Magnet-Polymer composites.* Macromolecular Chemistry and Physics, 2010. **211**(6): p. 618-626.
3. Nguyen Quang, V., *Magnet-Polymer composite transducers*, PhD thesis, School of Materials Science and Engineering, 2011, Nanyang Technological University: Singapore.
4. Snyder, R.L., V.Q. Nguyen, and R.V. Ramanujan, *Design of Magneto-elastic Soft Actuators.* Smart Materials and Structures, 2010. **19**(5): p. 055017-8.
5. R. Snyder, V.Q. Nguyen and R.V. Ramanujan, The energetics of magnetoelastic actuators is analogous to phase transformations in materials, *Acta Mater*., 58, 5620-5630 (2010)
6. A. S. Ahmed, V.Q. Nguyen and R. V. Ramanujan. *Multifunctional Smart Magnet-Polymer Composites (Magpol),* in International Symposium for Research Scholars on Metallurgy, Materials Science and Engineering, India (2012)
7. R. V. Ramanujan, *Intelligent magnet-polymer composite transducers (MAGACT)*, Plenary Presentation, in Nano 2011, Cochin, India (2011)
8. R. V. Ramanujan, V.Q. Nguyen and R. Snyder, *MAGACT: High performance smart active magnet-polymer composite transducers*, Invited Presentation, in ICMAT, Singapore (2011).
9. R. V. Ramanujan. and V. Q. Nguyen. *Magnet-Polymer Soft Transducers*. in MRS Spring Meeting, USA. (2010)
10. R. V. Ramanujan. and V. Q. Nguyen, *Magnet-Polymer Nanocomposites for Shape Changing Structures* (invited talk). in TMS Annual Meeting, USA. (2009).
11. V.Q. Nguyen and R. V. Ramanujan, *Mechanical Behavior of Novel Transducers Based on Magnetic-Polymer Composites* (invited talk). in ICMAT, Singapore. (2009).
12. V.Q. Nguyen and R. V. Ramanujan, *Magnet-Polymer composite material for intelligent shape changing structures*. in International Conference on Materials Processing for Properties and Performance, Singapore. (2008).
13. V.Q. Nguyen and R. V. Ramanujan, *Magnet-Polymer (MAGPOL) composite transducers for biomedical applications.* in International Conference on Cellular and Molecular Bioengineering, Singapore. (2007).