Axial strain of ferrogels under cyclic magnetic fields

L E Faidley\textsuperscript{1}, Y Han\textsuperscript{2}, K Tucker\textsuperscript{1}, S Timmons\textsuperscript{3} and W Hong\textsuperscript{2,3}

\textsuperscript{1} Mechanical Engineering Department, Iowa State University, Ames, IA 50011, USA
\textsuperscript{2} Aerospace Engineering Department, Iowa State University, Ames, IA 50011, USA
\textsuperscript{3} Materials Science and Engineering Department, Iowa State University, Ames, IA 50011, USA

E-mail: faidley@iastate.edu and whong@iastate.edu

Received 12 January 2010, in final form 22 April 2010
Published 21 May 2010
Online at stacks.iop.org/SMS/19/075001

Abstract

Ferrogels are low stiffness polymer materials with embedded magnetic powder filler, giving them the capability to strain in a magnetic field. The large strains, high energy densities and fast responses that have been reported make these materials attractive for actuator applications; however, a full understanding of the dynamic behavior is lacking. This paper presents an experimental study of the cyclic behavior of these materials under various frequencies in both the upright and inverted configurations. A 1D phenomenological magneto-viscoelastic model is developed and used to capture this behavior. The trends in stiffness, viscosity and density are described and then used to predict the amplitude of the strain at different frequencies.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

A ferrogel is a composite material comprised of a soft gel matrix and a filler of magnetic powder. These materials are magnetically activated smart materials with the capability to change their shape \cite{1, 2}, water retention \cite{3–5} and stiffness \cite{6, 7} when exposed to a magnetic field. Previous work by the authors \cite{8} and others \cite{2} has shown strains of up to 40\% when exposed to fields of up to 2500 G. These materials are also related to magnetorheological elastomers which have shown strains of about 16\% in uniform fields of 0.8 T \cite{9} and tens to hundreds of per cent in non-uniform fields \cite{10–12}. In addition, ferrogels have also shown load capabilities of up to four times the sample weight \cite{8}, translating into energy densities of about 400 J m\textsuperscript{-3} \cite{13}. Such observations suggest ferrogels for actuator behavior; however, before target applications can be chosen and actuators designed, the dynamic behavior of these materials needs to be better understood. This is the contribution of this work.

The mechanism for shape change in ferrogels lies in the magnetic force that governs the behavior of the magnetic particles in a field and the interaction of those particles with the soft gel matrix in which they are embedded. There are two contributions to the effect of the magnetic field on ferrogel samples \cite{1}. The dominant effect is the body force that creates forces on the magnetic particles due to a field gradient. The second is a surface force that is due to demagnetization effects and that causes deformation in ferrogels in uniform fields where the body force is zero.

The behavior of ferrogels in uniform fields has been studied extensively both theoretically \cite{1, 14, 15} and experimentally \cite{16}. It was found that spheres of ferrogel can strain up to 2\% when placed in a homogeneous magnetic field and form axisymmetrical non-spherical shapes with their axis aligned with the field direction. The strain mechanism is governed by balancing the Hooke elasticity with the Langevin magnetization laws.

The focus of this work is on the behavior as a result of a magnetic field gradient. The strain mechanism is illustrated in figure 1. When no field is present the monodomain particles have randomly oriented magnetization vectors. When a non-uniform field is applied the particles align with the field and move towards the area with the highest field intensity (i.e. the surface of the magnet). Because the matrix material is highly flexible the matrix moves with the magnetic particles, causing bulk strain. The magnetic attraction is counteracted by the internal friction of the viscoelastic matrix and, in some cases, the weight of the sample itself.

Zrinyi \textit{et al} did some early work focused on the behavior of ferrogels in non-uniform fields. In this work, ferrogels of PVA crosslinked with glutardialdehyde (GDA) and swollen
with a ferrofluid comprised of magnetic particles with an average size of 10 nm were synthesized and shown to strain in a magnetic field [17]. Cylindrical samples 1–2 cm in diameter and 10–20 cm long were exposed to fields of up to 0.8 T from an electromagnet perpendicular to the axis of the sample [2]. The field gradient needed for magnetic attraction of the particles is achieved by exposing only a portion of the sample to the field. Strains up to 40% and forces of 200 mN were measured. Preliminary basic modeling based on retarded elasticity and lumped inertia was performed [18]. This model was followed by more advanced modeling based on a force balance between magnetic and elastic forces on the nanoparticles within the sample that was done by Filipcsei et al [5].

One of the few dynamic studies of ferrogels was also conducted by Zrinyi et al [18]. In this work PVA samples crosslinked with GDA and swollen with a magnetic sol of Fe₃O₄ were tested at frequencies ranging from 0.1 to 100 Hz. They measured the frequency dependence of the initial susceptibility and the shape change. The susceptibility was found to be a constant across all frequencies and strain was achieved with little phase shift or relaxation up to 40 Hz. The work presented in this paper will confirm the findings of Zrinyi for a different ferrogel composition and field alignment and develop a viscoelastic model to capture this behavior.

2. Experimental approach

2.1. Sample preparation

The samples of ferrogel characterized during this work were prepared by chemically crosslinking poly(vinyl) alcohol (PVA) using sodium tetraborate. Sodium tetraborate crosslinks chains of PVA by linking oxygen ions through the borate ion as is shown in figure 2.

Samples were produced by adding 4 wt% PVA and 2 wt% sodium tetraborate in their powdered forms to 82 wt% distilled water creating the ionic bonds shown in figure 2. As the powders were being mixed with the water, 12 wt% carbonyl iron powder (Fe, from ISP Technologies, Inc. with an average diameter of 8.9 μm) was added to the solution. Stirring was continued until crosslinking was complete and a gel was formed. This resulted in a sample with homogeneously dispersed filler powder (as can be seen in figure 3), with a density of about 1700 kg m⁻³, determined by measuring the mass of a sample of known volume and a quasi-static elastic modulus of the order of 5 MPa. This particular composition was chosen as a continuation of the authors’ previous work [19] in which this composition was found to be the most active of the samples tested. It should be emphasized that the use of microscale powder added to the ferrogel during crosslinking varies substantially from other common methods of ferrogel synthesis that are found in the literature [17] and was chosen because it is substantially simpler than other methods and the micro-size particles have substantially fewer problems with agglomeration.

2.2. Dynamic testing apparatus

The set-up used to test the dynamic properties of the ferrogels is shown in figure 4(a). The magnetic field is produced by a solenoid made of eight layers of 100 turns each of 20 gauge copper magnet wire wound around a 3.8 cm diameter, 9 cm long 1018 low-carbon steel core. This solenoid has a resistance of 3.4 Ω and an inductance of 0.05 H and produces 153 G A⁻¹ on the surface of the core. Sinusoidal currents with a magnitude of 9 A were provided by a high current amplifier (AE Technon 7782) with a gain of 20 V/V which was driven by an NI cDAQ-9172 data acquisition system with a 9263 output and 9215 input module. Voltage inputs to the amplifier were calculated for each frequency to account for the inductance effects, and to keep the current amplitude—and therefore the field amplitude—constant. The strain output from the sample
was measured by way of a laser displacement sensor (Micro-Epsilon optoNCDT 1401) with a resolution of 0.6 μm and a measurement range of 5 mm. The sample itself was shaped into a 20 mm long, 10 mm diameter cylinder. It was placed on a 20 gauge copper alignment wire so that the alignment could be maintained with minimum constraint to the bulging of the sample base that is integral to the sample’s strain.

The apparatus shown in figure 4 can be set up with the two sample configurations shown in (b) and (c). In the upright configuration in (b) the sample rests on the solenoid core and the attractive force due to the field is in the same direction as gravity. In the inverted configuration in (c) the sample is reversed such that it is suspended below the solenoid core and gravity and the magnetic attraction oppose each other. In this orientation friction between the sample and the alignment wire, as well as the slight adhesiveness of the sample, hold the sample in contact with the solenoid core. Comparing tests run in the two configurations will give an indication of the effect of gravity on the ferrogel sample.

The field distribution predicted by a basic FEM magnetic model of the solenoid in the set-up in figure 4 is shown in figure 5. The amplitude of the field is 50% lower at the top surface of the sample area than it is at the top surface of the core. Though the distribution is not static and changes as the sample changes shape due to the magnetic properties of the ferrogel, it can be seen that a field gradient needed to achieve strains in the sample is produced.

### 2.3. Testing matrix

A series of tests were run in order to develop an understanding of how ferrogels react to dynamic magnetic fields and to inform the development of the analytical model. All of the tests were run on a ferrogel with a composition of 4% PVA, 12% Fe from the same batch. For each test a fresh sample was formed and mounted in the set-up, thus reducing variability in initial and boundary conditions and sample drying. Strain data was collected for both upright and inverted sample configurations at a variety of frequencies between 0.1 and 40 Hz.

### 2.4. Experimental results—cyclic strain

Tests were run as described in section 2.3 and a representative set of raw data is shown in figure 6 for 0.2 Hz. The zero time on the graphs represents the start of the data collection, which occurred after the sample had reached steady state. Thus, this data does not include any instantaneous effects. The shape of this curve is a sinusoidal strain superimposed on a linear decrease (upright orientation) or increase (inverted orientation) which is expected for a viscoelastic material exposed to both gravity and a cyclic excitation. A negative strain represents the compression of the sample while a positive strain is the elongation of the sample. The frequency of the strain is double that of the input field since both a large negative field and a large positive field cause the sample to strain towards the magnet. There is also a slight phase shift seen between input and output due to the inductance of the driving coil.

The slopes and cyclic strain amplitudes for tests run at a variety of frequencies and in both orientations are shown in figure 7. In figure 7(a) the amplitude of the cyclic strain for the inverted and upright configurations are shown along with the averages of these numbers. As expected, the amplitude of the cyclic strain is found to be very similar for both orientations as gravity does not influence this quantity. The larger strains at lower frequencies that plateau at higher frequencies are also expected for this viscoelastic material as the rate of excitation determines the amount and type of strain. It is believed that the 40 Hz outlier seen in this data is caused by resonances within the test stand that are excited at this high frequency. This point is ignored in the calculations shown in section 3.3. The other smaller discrepancies seen in this data are an effect of the difficulties that exist in establishing consistent initial and boundary conditions when working with soft polymeric materials like ferrogels.
In figure 7(b) the slope values for each frequency are shown for the upright (red square) and inverted (blue diamond) configurations. This graph shows a negative (compressive) slope for the upright case where both gravity and the magnetic force act to compress the sample and a positive (tensile) slope for the inverted case where the magnetic compressive effect is counteracted by a tensile gravity effect. The relative magnitudes of these slopes also reflect these additive and counteractive effects. The solid lines represent the slope for each configuration measured under zero field and thus capture the gravity effect on the sample. When a field is applied the slope decreases due to the magnetic force which is always compressive. Since the field applied has a constant amplitude in these tests, the slope seen can be approximated as a constant.

3. Analytical development

3.1. Qualitative prediction

Let us begin by examining the behavior of a ferrogel sample in a static magnetic field. The two forces of interest are (1) gravity, which causes deformation of the sample due to the viscoelastic properties of the matrix and (2) the magnetic body force, which causes deformation of the sample due to the attraction of the magnetic filler to the region of highest magnetic field. Thus, in the upright case the static strain will be larger since gravity and the magnetic force act in the same direction; both causing the sample to become shorter. In the inverted orientation, however, the gravity acts down on the sample, causing it to become longer, and the magnetic force acts upwards, causing it to become shorter. The balance of these two forces results in smaller static strains for inverted cases than for the upright case.

Now let us consider the behavior of a ferrogel sample under the cyclic application of a magnetic field. In this situation we expect two components to be evident in the strain

Figure 6. Representative experimental data at 0.2 Hz showing (a) input field, (b) upright strain response and (c) inverted strain response.

Figure 7. Experimental determination of (a) cyclic strain amplitude ($A$) and (b) slope for upright and inverted configurations ($m$).
of the sample. First is the continuous lengthening (inverted) or shortening (upright) of the sample due to its viscoelastic behavior under gravity and the applied field. Superimposed on this continuous strain will be a cyclic strain that is driven by the application of the cyclic field and the internal elastic restoring forces of the sample. This description agrees with the strains observed experimentally as shown in figure 6. The overall slope of this curve is caused by gravity acting on the sample and will be determined by the internal viscosity of the sample as well as its size and geometry and specific weight. The cyclic part of the strain will occur at double the frequency of the applied field since the particles will be attracted towards the magnet for both large negative and large positive fields.

The amplitude of the cyclic strain is caused by the attractive forces between the field and the magnetic filler, causing motion towards the region of highest field. When the field is removed the internal elasticity of the sample forces it to return towards its original shape, creating cyclic strain. The magnitude of this cyclic strain will depend on the size of the field, the volume fraction of the magnetic filler and the internal elasticity of the sample.

3.2. Quantitative model

To quantify the discussion in section 3.1 we recognize two sources of the body force in the ferrogels: the gravity-generated body force \( b_g \) and the magnetic body force \( b_m \). While \( b_g \) scales with the specific weight of the sample, \( \rho g \), the magnetic contribution is

\[
b_m = \mathbf{M} \cdot \nabla \mathbf{B},
\]

where \( \mathbf{M} \) is the magnetic dipole moment per unit volume and \( \mathbf{B} \) is the local magnetic induction, both being spatially inhomogeneous. A profile of the magnetic induction field is shown schematically in figure 5. Without further knowledge of the magnetic properties of the composite, we assume that both \( \mathbf{M} \) and \( \mathbf{B} \) are linearly proportional to the local magnetic field, \( \mathbf{H} \).

Neglecting the constraints and the boundary effects, we assume the samples to be in a statically determinate state, so that the stress can be evaluated from a direct integration of the body forces:

\[
\sigma = \pm \rho g z - f(z)H^2,
\]

where \( f(z) \) is a function determined by the spatial distribution of the magnetic field and the magnetic permeability of the material, and \( H \) is a scalar measurement of the external magnetic field. The positive sign in the first term in equation (2) is associated with the inverted orientation and the negative sign corresponds to the upright set-up. We assume the deformation to be small enough that the spatial profile of the magnetic field is not affected. Our experimental set-up dictates that the magnetic-field-induced stress to be always compressive, while the gravity-induced stress is compressive for the upright case and tensile for the inverted case.

In this paper, we are looking at the low-frequency response of ferrogels, and the inertia effect is thus neglected. By adding a term from the inertial force in equation (2), the model can be easily extended to account for the inertia effect at a relatively high frequency.

Inspired by the qualitative predictions described above and the experimental observation shown in figure 6 we adopt the Maxwell spring-dashpot model as sketched in figure 8, with the following stress–strain relation:

\[
\sigma = k \epsilon_e + \eta \dot{\epsilon}_e.
\]

Here \( \epsilon_e \) and \( \epsilon_v \) represent the elastic and viscous parts of the strain, respectively. The strain contributions are additive and the total strain is \( \epsilon = \epsilon_e + \epsilon_v \).

In the experiment, a sinusoidal magnetic field was applied with the form

\[
H = H_0 \sin(\omega t).
\]

Substituting equations (2) and (4) into (3), after integrating the total strain over the length of the sample, \( L \), we obtain the average strain of the whole sample:

\[
\bar{\epsilon} = \frac{1}{k} \left( \pm \rho g L - \frac{p H_0^2}{2} \right) + \frac{p H_0^2}{2} \left( \frac{1}{k} \right)^2 + \frac{1}{2 \omega \eta} \sin(2 \omega t \pm \phi),
\]

where \( \phi = \arctan \left( \frac{2 \omega \eta}{k} \right) \) and \( p \) is a positive scaling constant that depends on the permeability of the material. The average strain in equation (5) consists of three parts, the instantaneous strain:

\[
\epsilon_i = \frac{1}{k} \left( \pm \rho g L - \frac{p H_0^2}{2} \right),
\]

the ramped strain with a slope \( m \) of

\[
m_{\pm} = \frac{1}{\eta} \left( \pm \rho g L - \frac{p H_0^2}{2} \right),
\]

and the oscillatory strain with an amplitude of

\[
A = \frac{p H_0^2}{2} \sqrt{\left( \frac{1}{k} \right)^2 + \left( \frac{1}{2 \omega \eta} \right)^2},
\]

a frequency of \( 2 \omega \) and a phase of \( \arctan \left( \frac{2 \omega \eta}{k} \right) \). Note that the slope can be either positive or negative, depending on the orientation of the sample and the relative magnitudes of the specific weight and the magnetic force. The amplitude is independent of gravity and thus is the same for both orientations. This confirms the observations made in section 2.4. No observation was made of the instantaneous strain since testing began at steady state in the current experiment.

From the experimentally measured strain output, we can extract the slopes of the ramped strain in both the upright and
Figure 9. Experimental results and fitted model parameters: (a) $\alpha$, (b) $\beta$ and (c) $\gamma$. (d) Experimental results and model prediction of the oscillatory strain amplitude, as a function of the frequency of magnetic field ($A$).

Of these, $\alpha$ and $\gamma$ are dimensionless while $\beta$ bears the dimension of time. While all of them are affected by the magnetic permeability of the material and the strength of the applied field, $\alpha$ measures the stiffness of the ferrogel, $\beta$ measures the viscosity and $\gamma$ measures the specific weight.

3.3. Model results and improvement

Using the measured strain of samples from the same batch of ferrogel, we extract the three material parameters, $\alpha$, $\beta$ and $\gamma$, and plot them against the frequency of the applied field in figures 9(a)–(c). Within the accuracy of measurements, the values of both $\beta$ and $\gamma$ remain constant throughout the frequency range, while that of $\alpha$ increases with frequency below 1 Hz and then remains constant. A fitting expression of the form $\alpha = \frac{k}{pH_0}$, $\beta = \frac{\eta}{pH_0}$, and $\gamma = \frac{\rho g L}{pH_0}$.

Of these, $\alpha$ and $\gamma$ are dimensionless while $\beta$ bears the dimension of time. While all of them are affected by the magnetic permeability of the material and the strength of the applied field, $\alpha$ measures the stiffness of the ferrogel, $\beta$ measures the viscosity and $\gamma$ measures the specific weight.

3.3. Model results and improvement

Using the measured strain of samples from the same batch of ferrogel, we extract the three material parameters, $\alpha$, $\beta$ and $\gamma$, and plot them against the frequency of the applied field in figures 9(a)–(c). Within the accuracy of measurements, the values of both $\beta$ and $\gamma$ remain constant throughout the frequency range, while that of $\alpha$ increases with frequency below 1 Hz and then remains constant. A fitting expression of the form $\alpha = \frac{k}{pH_0}$, $\beta = \frac{\eta}{pH_0}$, and $\gamma = \frac{\rho g L}{pH_0}$.

4. Conclusions

In this paper we have presented a study of the strain behavior of ferrogel samples under cyclic magnetic excitation. Experimental testing measured the displacement of a 12 wt% Fe, 4 wt% PVA sample when exposed to a cyclic field with an amplitude of 2575 G in both an upright and inverted configuration. The measured strain is a sinusoid, with a frequency double that of the applied field, superimposed on a linear function, with the sign of the slope determined by the relative directions of the magnetic force and gravity. The form of this strain suggests a Maxwell viscoelastic model.
of the ferrogel sample comprised of an effective stiffness and viscous element in series. This model was developed and three parameters, $\alpha$, $\beta$, and $\gamma$, dependent on stiffness, viscosity and density, respectively, were derived. Experimental determination of the slopes and cyclic strain amplitudes under both inverted and upright sample configurations at frequencies from 0.1 to 40 Hz allowed for $\alpha$, $\beta$ and $\gamma$ to be found. $\beta$ and $\gamma$ are nearly frequency-independent and $\alpha$ demonstrates a weak exponential dependence on frequency, typical of gels. A model improvement eliminates the frequency dependence of $\alpha$ by adding two elements to the Maxwell model and using five parameters to capture the sample behavior. The model was able to capture the cyclic strain amplitude dependence on frequency for this particular sample. It is expected that similar models with different parameters will be able to predict the behavior of other ferrogel compositions. Future work in this area will capture the physical changes generated by these composition changes and how they affect the model parameters, moving towards a more generic model for these materials.

**Acknowledgments**

The authors would like to acknowledge the financial support of the National Science Foundation grant no. 0900342. We also thank Mary Burroughs and David Macias for their help with data acquisition.

**References**


---

**Figure 10.** Four-element viscoelastic model. (a) The experimental data of the cyclic strain amplitude and the model prediction. Inset: a sketch of the model used. (b) The slope of the measured slopes of the ramped strains, in the upright and inverted set-ups, and the model predictions.