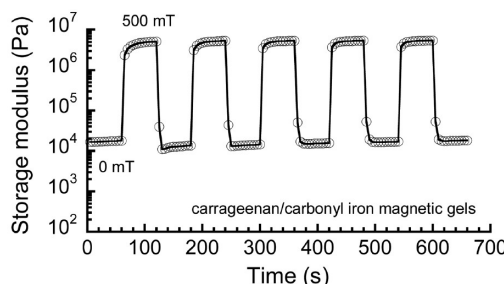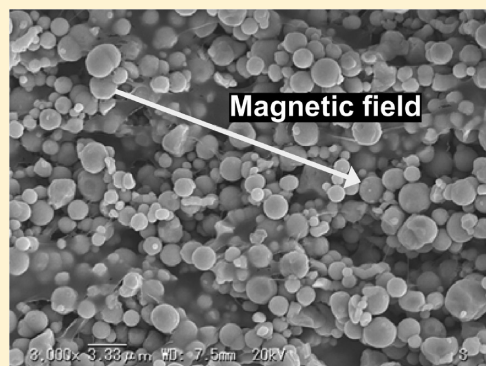# Magnetically Tunable Elasticity for Magnetic Hydrogels Consisting of Carrageenan and Carbonyl Iron Particles

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ABSTRACT: A new class of magnetoelastic gel that demonstrates drastic and reversible changes in storage modulus without using strong magnetic fields was obtained. The magnetic gel consists of carrageenan and carbonyl iron particles. The magnetic gel with a volume fraction of magnetic particles of 0.30 exhibited a reversible increase by a factor of 1400 of the storage modulus upon a magnetic field of 500 mT, which is the highest value in the past for magnetorheological soft materials. It is considered that the giant magnetoelastic behavior is caused by both high dispersibility and high mobility of magnetic particles in the carrageenan gel. The off-field storage modulus of the magnetic gel at volume fractions below 0.30 obeyed the Krieger–Dougherty equation, indicating random dispersion of magnetic particles. At 500 mT, the storage modulus was higher than 4.0 MPa, which is equal to that of magnetic fluids, indicating that the magnetic particles move and form a chain structure by magnetic fields. Morphological study revealed the evidence that the magnetic particles embedded in the gel were aligned in the direction of magnetic fields, accompanied by stretching of the gel network. We conclude that the giant magnetoelastic phenomenon originates from the chain structure consisting of magnetic particles similar to magnetic fluids.

1. INTRODUCTION

Biological tissue, such as the dermis of sea cucumbers, exhibits stimuli-responsive properties where the elasticity alters in response to a physical stimulus. Materials where the physical property changes in response to a stimulus have been extensively investigated, particularly in the field of soft materials, such as polymer gels, rubbers, or elastomers. It is natural to consider that polymer gels are especially suitable for fabricating stimuli-responsive materials because polymer gels consist of a large amount of water, similar to biological tissue. It is because materials containing fluids have a high degree of freedom, such as macroscopic deformation, movement of network or solute, etc. So far, many stimuli-responsive polymer gels sensitive to pH, solvent composition, temperature, electric field, magnetic field, and ultrasounds have been reported.

Polymer gel containing magnetic particles is a stimuli-responsive gel where its viscoelastic properties can be controlled by applying magnetic fields. There have been many attempts to fabricate magnetoelastic materials using soft materials, such as synthetic polymer gels,[1] silicone elastomers,[2−7] and rubbers.[8,9] Zrinyi et al. performed pioneering work in this field and reported various magnetic gels demonstrating elongation behavior or variable elasticity. Details

of the past investigations about magnetoelastic gels and elastomers have been reviewed in the literature.[10−15] The elastic modulus of magnetic gels is enhanced by forming a chain structure of magnetic particles, similar to magnetic fluids. The stability of magnetic particles in magnetoelastic solids is superior to that in magnetoelastic fluids because of the high viscosity of magnetoelastic solids; for example, the sedimentation of magnetic particles does not take place easily in solids. However, the magnetoelastic response of solids was generally small compared with that of fluids. This originates mainly from the reason that the magnetic particles are difficult to move and form a chain structure in solids. In many cases for magnetoelastic solids, the relative change in the storage modulus (the ratio of storage moduli at magnetic fields to off-fields) was less than several times. However, significant progress can be seen recently in the development of soft materials using a magnetoelastic effect; for example, a highly swollen physical gel demonstrates the relative change in the storage modulus of 60 times.[16]
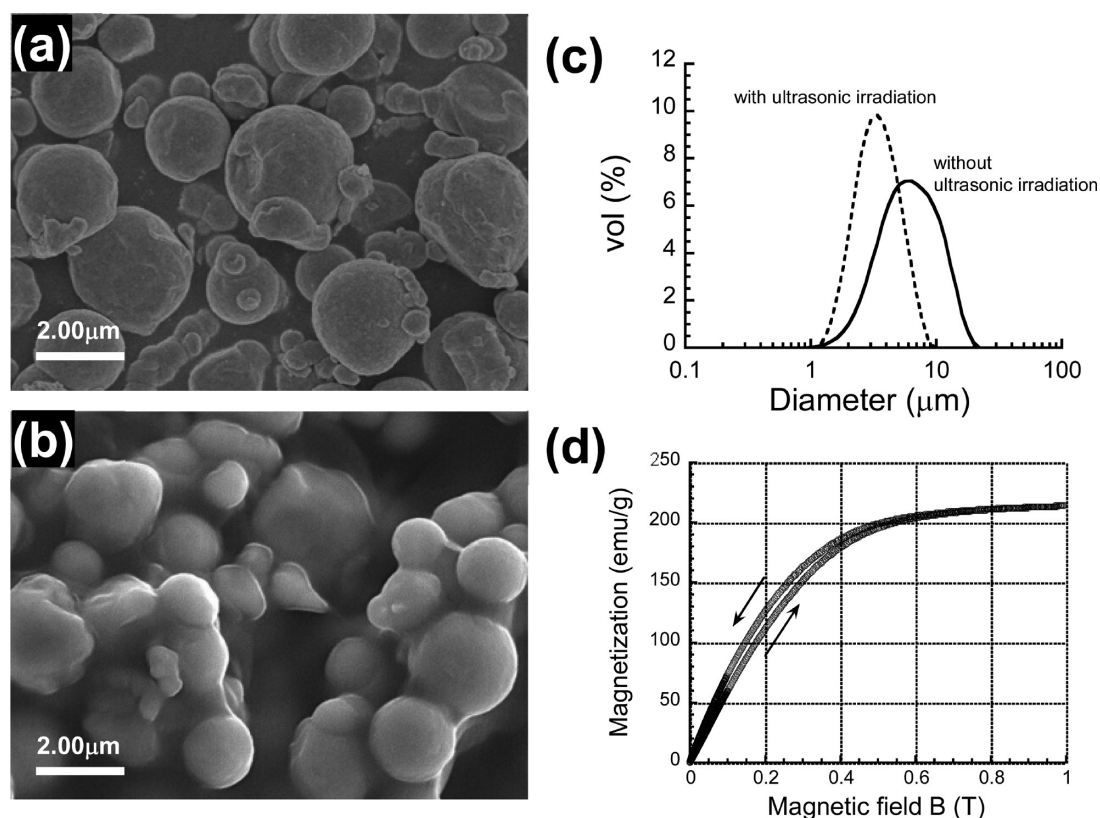
Figure 1. SEM photographs for carbonyl iron (CI) particles (a) before and (b) after being embedded in carrageenan (CA) gel. (c) Particle diameter distribution and (d) magnetization curves for the CI particles used in the present study.

A few years before, we succeeded in fabricating a new class of magnetoelastic gel that demonstrates drastic and reversible changes in the dynamic modulus without using strong magnetic fields.[17] At zero magnetic field, the storage modulus of the magnetic gel is extremely low, $\sim 10^4$ Pa, although the gel contains a large amount of particles as much as $\phi \sim 0.30$, resulting from random dispersion of the magnetic particles in the gel. The magnetic particles, under a magnetic field, align to the magnetic lines of force and form a chain structure contributing to the high storage modulus exceeding 4 MPa. In 2011, we reported a success for synthesizing polyurethane elastomers showing drastic change in the dynamic modulus by a factor of 277.[18]

In this paper, we present the magnetic, viscoelastic, and morphological properties of magnetic carrageenan gel that underwent a wide modulation of the dynamic modulus (500 times higher than off-field modulus).[17] In addition, we show new evidence that magnetic carrageenan gels exhibited a huge increase in storage modulus (1400 times higher than off-field modulus) by decreasing the carrageenan concentration. The mechanism of the giant magnetoelastic behavior demonstrated by magnetic carrageenan gels is discussed.

2. EXPERIMENTAL PROCEDURE

Synthesis of Magnetic Elastomers. The magnetic gel consisted of carbonyl iron (CI) particles (BASF, Japan) and κ-carrageenan (M_w = 857 kDa, CS-530, San-Ei Gen F.F.I.). A pregel solution of the magnetic gel was prepared by mixing a 1.0 wt % carrageenan aqueous solution and CI particles at 95 °C using a mechanical stirrer for 30 min. The diameter of the primary particle was 2.5 μm, and the saturation magnetization

and coercive force were 214 and 1.1 emu/g, respectively. The particle has no magnetization at synthesis; it was magnetized under magnetic fields. The weight concentration of CI particles was varied from 0 to 73 wt %, which corresponds to a volume fraction of 0−30 vol %. The volume fraction of magnetic particles was calculated using the following equation:

$$\phi = \frac{d_{MG} - d_{CA}}{d_{MP} - d_{CA}} \tag{1}$$

Here, d_{MG} denotes the density of the magnetic gel, d_{CA} is the density of the carrageenan gel as a matrix, and d_{MP} is the density of the magnetic particles. The measured density of the carrageenan gel and CI particles used in this study was 1.00 and 6.29 g/cm^3, by Archimedes' principle.

Magnetic Measurements. The magnetic measurement for the CI particles and magnetic elastomers with various volume fractions was carried out at 298 K by using a vibrating specimen magnetometer (Tamakawa Seisakusho, Ltd.).

Rheological Measurements. Dynamic viscoelastic measurements were carried out using a rheometer (MCR301, Anton Paar) with a nonmagnetic measuring plate, which is a special plate for the measurement under a magnetic field. For strain sweep measurements, the frequency was kept at 1 Hz, and the strain amplitude was varied from 10^{-6} to 10^{-2}. For the measurement of the magnetic field response, the frequency and strain were kept at 1 Hz and 10^{-4}, respectively. The temperature was controlled at 20.0 °C during the viscoelastic measurement. The samples were disks with dimensions of 20 mm in diameter and 1.5 mm in thickness.

SEM Observations. The shape of the CI particles was observed using a scanning electron microscopy (SEM) with an

accelerating voltage of 20 kV (VE-9800, Keyence). The CI particles showed a spherical shape (Figure 1a), and the diameter of the CI primary particle was determined to be 2.5 ± 0.2 μm by a particle size analyzer (SALD-7000, Shimazu), as shown in Figure 1c. SEM photographs of the magnetic gel for random dispersion were taken after freeze-drying for 10 h without applying magnetic fields. To observe the chain structure of magnetic particles, the magnetic gel was freeze-dried for 10 h under a magnetic field of 320 mT using a permanent magnet. Apparent shrinkage was not observed for both cases due to the freeze-drying (less than 3% in length).

3. RESULTS AND DISCUSSION

Figure 1a,b shows the SEM photographs for magnetic particles before and after being embedding in carrageenan gel, respectively. The photograph of magnetic particles before embedding in the gel indicates the fact that each particle is isolated individually and has a metallic rough surface. After the particles were embedded in the gel, it was observed that the particles were coated with a thick layer of carrageenan, which was bound on the magnetic particles, and the particles were seen to be connected to each other. This indicates a feature that the carrageenan has adhered against the surface of the magnetic particles. Figure 1c illustrates the distribution profile of the particle diameter for the magnetic particles used in the present study. The mean diameter of magnetic dispersion just after irradiation of ultrasound was determined to be 3.2 μm, which equals to that of the primary particle measured in air (=2.5 μm). The dispersion without irradiation of ultrasound showed a broad distribution of particle diameter, and the mean diameter shifted to a high value of 8.0 μm. This means that the magnetic particles form secondary particles in pure water. When the magnetic particles were dispersed in a dilute solution of carrageenan, no sedimentation was observed for several weeks. This means that the carrageenan behaves as a dispersing agent. Figure 1d displays the magnetization curves for the magnetic particles (powder state) used in the present study. The magnetization increased with increasing the magnetic field and saturated at around 0.8 T. Note that the viscoelastic measurement was carried out at 500 mT, with the magnetization values of ~91% of the full moment of the magnetic particles. Regarding the process of demagnetization, the values of magnetization were higher than those of the magnetizing process; however, the remanent magnetization was 1.1 emu/g, suggesting that the magnetic moment disappeared after removing the magnetic field, which is small enough to be neglected.

Figure 2 demonstrates the strain dependence of the storage modulus G' for CA/CI magnetic gels in the presence and absence of magnetic fields. The storage modulus of the carrageenan gel without magnetic particles was ~3.2 × 10^3 Pa in the linear viscoelastic region. As the volume fraction of magnetic particles was increased, the storage modulus at the linear viscoelastic region increased. Simultaneously, the magnetic gels with high volume fractions exhibited nonlinear viscoelasticity at high strains, which has been called the Payne effect.[19] We have reported similar behavior seen in magnetic gels and elastomers with high volume fractions of magnetic particles.[20,21] It is worth mentioning that the magnetic gel containing CI particles exhibits weak nonlinear viscoelasticity compared to iron oxide (Fe$_3$O$_4$) or barium ferrite (BaFe$_{12}$O$_{19}$). This strongly indicates a feature that the CI particles disperse without making large clusters in the carrageenan gel, as
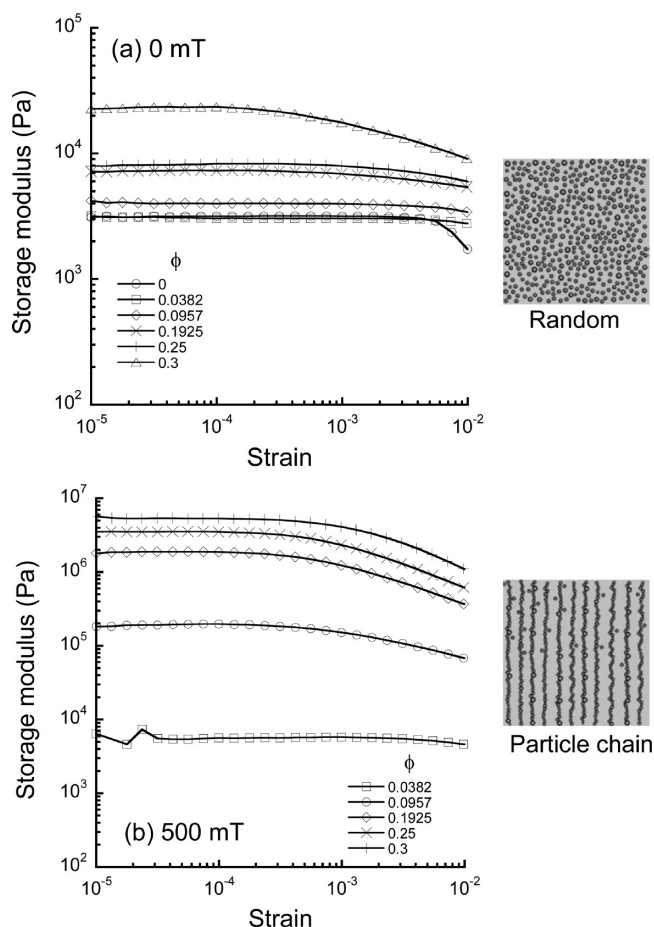
Figure 2. Strain dependence of storage modulus for CA/CI magnetic gels at (a) 0 mT and (b) 500 mT for various volume fractions of magnetic particles.

schematically illustrated in Figure 2a. When a magnetic field of 500 mT was applied, the storage moduli of these magnetic gels dramatically increased. It was also observed for all volume fractions that the nonlinear viscoelasticity was enhanced compared with those at 0 mT, meaning the number of particles making contact increased by applying a magnetic field, as illustrated in Figure 2b.

Figure 3a shows the volume fraction dependence of the storage modulus G' for CA/CI magnetic gels in the presence and absence of a magnetic field. The storage modulus represents the values in the viscoelastic regime in Figure 2. The solid line in the figure represents the storage modulus calculated by the following Krieger–Dougherty equation for a random dispersion of particles[22]

$$G' = G_m' \left(1 - \frac{\phi}{\phi_m}\right)^{-5/2\phi_m}$$

(2)

where G' and G'_m are the storage moduli of the magnetic gel and carrageenan gel, respectively. ϕ_m is the maximum volume fraction of the magnetic particles, which is approximately 0.63 for hard spheres.[23] At 0 mT, the storage modulus of the magnetic gel almost obeyed the above equation up to $\phi \sim 0.33$, indicating that the magnetic particles are homogeneously dispersed in the carrageenan gel. We have reported that the CI particles show high dispersibility also in polyurethane elastomers.[18] This causes a giant and reversible change in the
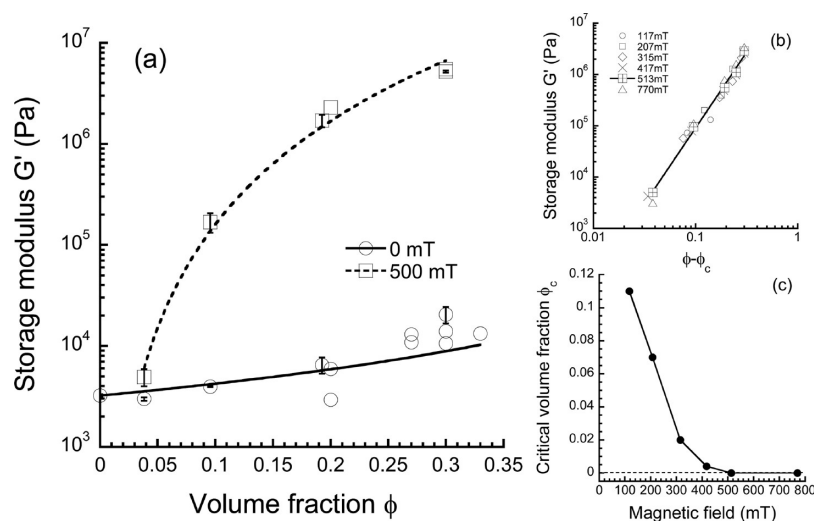
Figure 3. (a) Volume fraction dependence of storage modulus in the linear viscoelastic regime for CA/CI magnetic gels at 0 and 500 mT. (b) Storage modulus in the linear viscoelastic regime for CA/CI magnetic gels as a function of reduced volume fraction. (c) Critical volume fraction as a function of magnetic field.

elastic modulus of the magnetic elastomers. At 500 mT, the storage moduli for the magnetic gels were higher than those at 0 mT. When the volume fraction was $\phi < 0.04$, no significant increase in the storage modulus was observed upon an application of a magnetic field. The storage modulus at 500 mT can be well fitted by a power law (broken line), which is an indication of the percolation threshold at $\phi \sim 0.04$.

As shown in Figure 2, the storage modulus originates from the chain structure consisting of CI particles. It can be easily expected that the number of chains increases with either the volume fraction of CI particles or the magnetic field strength. Therefore, the storage modulus under magnetic fields should be described by a function of either of these parameters. Figure 3b illustrates the relationship between the storage modulus and the reduced volume fraction of magnetic particles. As seen in Figure 3a, the storage modulus of the magnetic gels at 500 mT demonstrated typical concentration percolation behavior. To determine the critical volume fraction ϕ_c and the critical exponent β, a power law is used

$$G' \propto (\phi - \phi_c)^\beta \qquad (3)$$

where G' and ϕ represent the storage modulus and the volume fraction of magnetic particles, respectively. After subtracting the contribution of ϕ_c, all data measured at various magnetic fields distributed on a straight line. The critical exponent β was determined to be 3.0 ± 0.2 by least-mean-squares fitting, which is very close to the value of 3d site percolation for a simple cubic.

Figure 3c displays the critical volume fraction ϕ_c obtained from the fitting by eq 3 as a function of magnetic fields. The value of ϕ_c was 0.11 at 100 mT, and it decreased remarkably with the magnetic field. This result suggests the evidence that a weak magnetic field (e.g., 100 mT) does not produce the macroscopic chain structure at $\phi < 0.11$, but a strong magnetic field (e.g., 500 mT) produces the chain structure even at extremely low volume fractions. We conclude that the magnetoelastic behavior of magnetic carrageenan gels originates from the formation of particle chains in carrageenan gel, which allows macroscopic stress transfer at volume fractions above ϕ_c.

Figure 4 displays the SEM photographs for CA/CI magnetic gels freeze-dried under a magnetic field with field strengths of 0 mT (left photos) and 320 mT (right photos). No alignment was observed for the CI particles when the magnetic field was 0 mT. It can be seen that the CI particles align in the direction of magnetic fields, which is called a chain structure (right photographs). This evidence clearly indicates that the magnetic particles are able to move within the carrageenan gel, which was cross-linked by the physical bond. We have already observed a phenomenon for carrageenan/γ-Fe$_2$O$_3$ hydrogels that the storage modulus was reduced remarkably by the particle movement.[24] This movement of magnetic particles must accompany the local deformation of the gel network since the magnetic particles are immobilized by the network. The shape of magnetic gels cannot be altered during the viscoelastic measurement because the gel is sandwiched between two parallel plates. The arrangement of magnetic particle changes from random to a chain structure without changing its shape macroscopically. Indeed, we found several traces that the carrageenan network was stretched in the direction of the magnetic field, as indicated by white arrows in Figure 4d and black arrows in Figure 4g. This evidence also suggests that there arises a strong interaction between magnetic particles and the carrageenan gel. Adhesive properties between particles and a matrix would be an important factor for the giant magneto-elasticity of magnetic gels.

Next, we estimate the moving distance of magnetic particles within the gel and compare with the particle size. Hypothesizing that one magnetic particle occupies the space volume with a distance between the nearest-neighbor particles, the distance between magnetic particles d can be roughly estimated by the following equation

$$d = 2r(\phi^{-1/3} - 1) \qquad (4)$$

where r and ϕ represent the radius and the volume fraction of magnetic particles. A schematic illustration representing the movement of magnetic particles is shown in Figure 5. When the volume fraction of the magnetic particles is 0.30, the distance between the nearest-neighbor particles is calculated to be 4.5 μm, which is shorter than the distance between two particles. The distance between the particles is quite short; therefore, the

Without magnetic field   **With magnetic field**
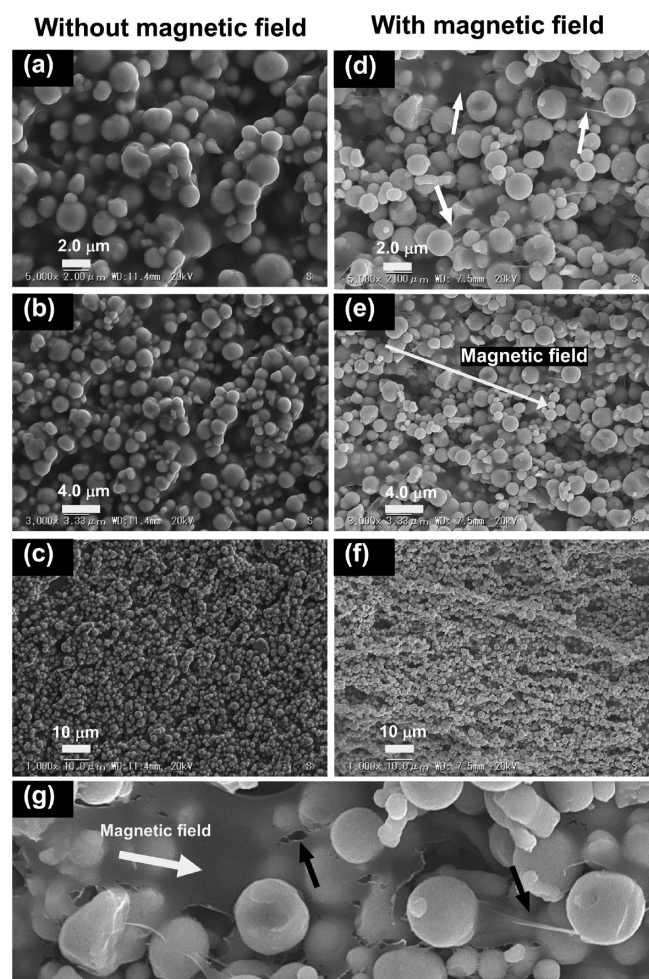
Figure 4. SEM photographs for CA/CI magnetic gels freeze-dried under magnetic fields of 0 mT (a−c) and 320 mT (d−f) (CI: 27 vol %). Arrows in (d) indicate the part where the carrageenan was stretched due to the chain formation. The bottom photo is an enlarged photo of (d). Black arrows indicate a ripped (left) part and the stretched (right) part by the chain formation.
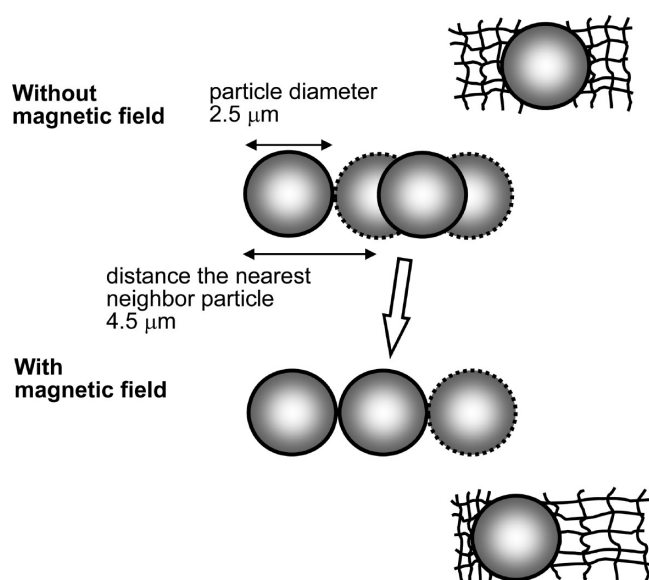

Figure 5. Schematic illustrations representing the movement distance of magnetic particles by magnetic fields.

magnetic particles are considered to make contact easily with neighboring particles. Under magnetic fields, the carrageenan network at the right side of the magnetic particle would be elongated; oppositely, the network at the left side would be compressed. Figure 6 shows the magnetic field response of the storage modulus in the linear viscoelastic regime for CA/CI magnetic gels. A magnetic field of 500 mT was applied to the magnetic gels every 60 s. It was observed that the storage modulus increased synchronized with the magnetic field and completely recovered to the original modulus after removing the field. The elongation and compression forces acting on the magnetic particles should contribute to the recoveries of the storage modulus. Therefore, the perfect recovery indicates that the elongational and compressional deformations occur within the limitation of elastic deformation, without causing the destruction of the polymer network.

Figure 7 demonstrates the magnetic field dependence of the storage modulus for CA/CI magnetic gels. The storage modulus of the gel increased with the magnetic field and was saturated at high magnetic fields. At first sight, the magnetic field profiles of the storage modulus resemble the magnetization curve of magnetic particles, as shown in Figure 1. This means that the magnetically induced modulus is attributed to the magnetic interaction between the magnetic dipole and the magnetic field. Generally, the Maxwell's stress is generated perpendicularly to the magnetization direction when a magnetic field is applied to a magnetic substance. The magnetically induced stress $\Delta G'$ is explained by the following equation[25]

$$\Delta G' \sim \mu_r \mu_0 (M\phi)^2 \tag{5}$$

Here, μ_r is the relative magnetic permeability of a bulk magnetic gel, which is the magnetic permeability with respect to that in vacuum μ_0 (=1.26 × 10^{-6} H/m). M and ϕ represent the magnetization of magnetic gels and the volume fraction of magnetic particles, respectively. Figure 8 shows the relationship between the absolute change in the storage modulus and the value of $M\phi$ calculated from the magnetization curve (Figure 1). The result of fitting by eq 5 is shown as broken lines in the figure. The $\Delta G'$ at the low region of $M\phi$ was well fitted by eq 5; however, at the high region of $M\phi$, it deviates from the fitting line and tends to saturate. After the saturation of the narrow region, the $\Delta G'$ significantly increased with the $M\phi$, indicating that the increase in the storage modulus is caused by another mechanism that is different from that at the low region of $M\phi$. The magnetic particles may form thicker chains to become a magnetically stable structure by the strong magnetic field, although it is now unclear. The relative permeability for these gels was calculated to be 4.55, 6.40, 10.4, 12.8, and 26.5 H/m, respectively. On the other hand, the value of μ_r can be calculated from magnetic data using the linear combination of the relative permeability of carrageenan, μ_{CA} (~1.0), and the CI particles, μ_{CI} (~5.48), that is, $\mu_r = \phi\mu_{CI} + (1 - \phi)\mu_{CA}$, since the magnetism of CI particles is not interfered by carrageenan gels of nonmagnetic materials. Using the linear combination, the magnetic permeability for these gels was calculated to be 1.2, 1.4, 1.9, 2.1, and 2.3 H/m, respectively. The magnetic permeability estimated by the relationship between $\Delta G'$ and $M\phi$ was apparently higher than the calculated values, indicating that the magnetically induced stress is mainly caused by the particle chain, not magnetic interaction between magnetic particles.
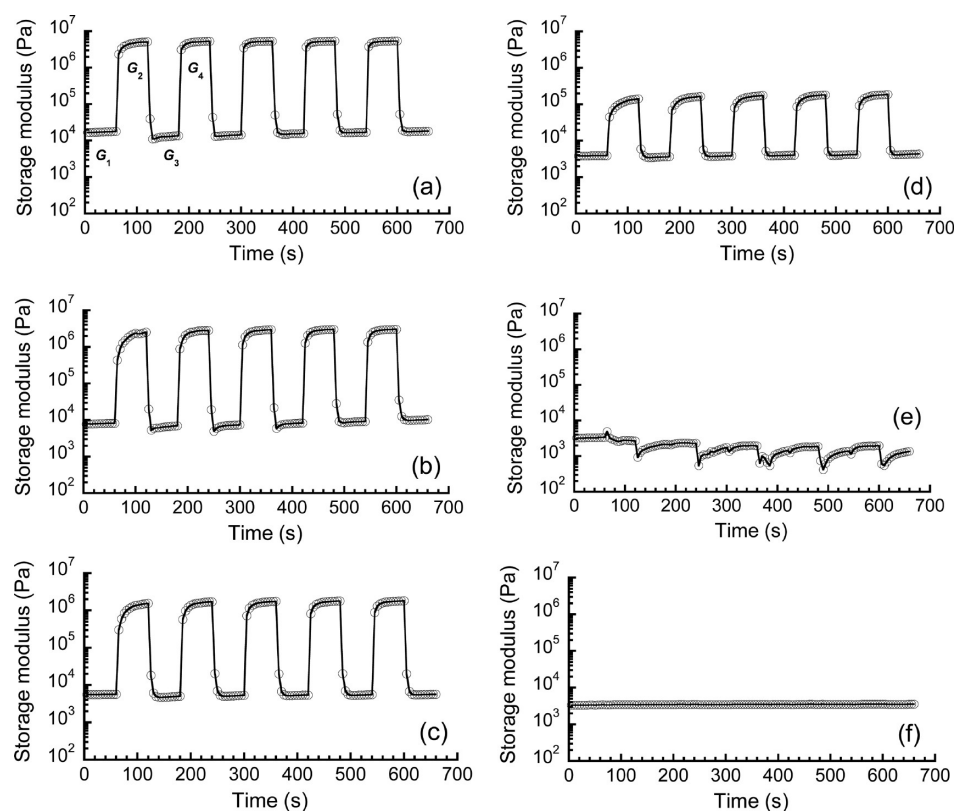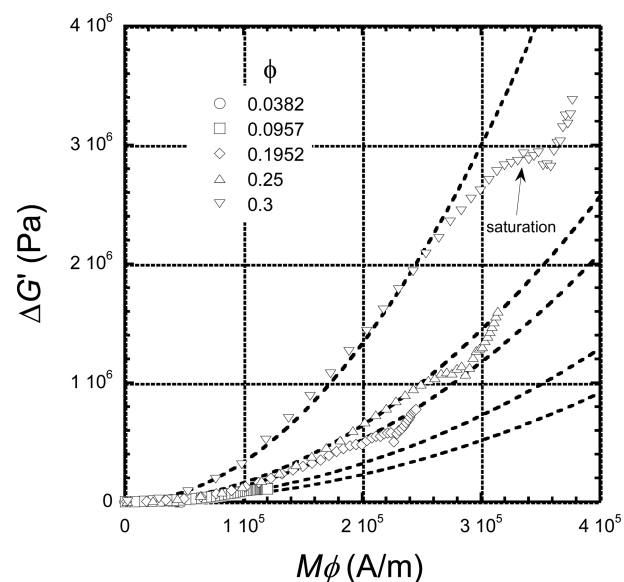
Figure 6. Magnetic field response of storage modulus for CA/CI magnetic gels with various volume fractions: (a) 0.30, (b) 0.25, (c) 0.19, (d) 0.10, (e) 0.038, and (f) 0.00.
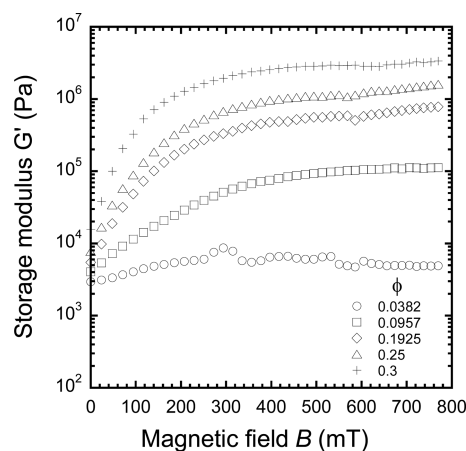
Figure 7. Magnetic field dependence of storage modulus in the linear viscoelastic regime for CA/CI magnetic gels with various volume fractions of magnetic particles.

Figure 8. Absolute change in storage modulus $\Delta G'$ as a function of $M\phi$ represented in eq 5 for various volume fractions of magnetic particles.
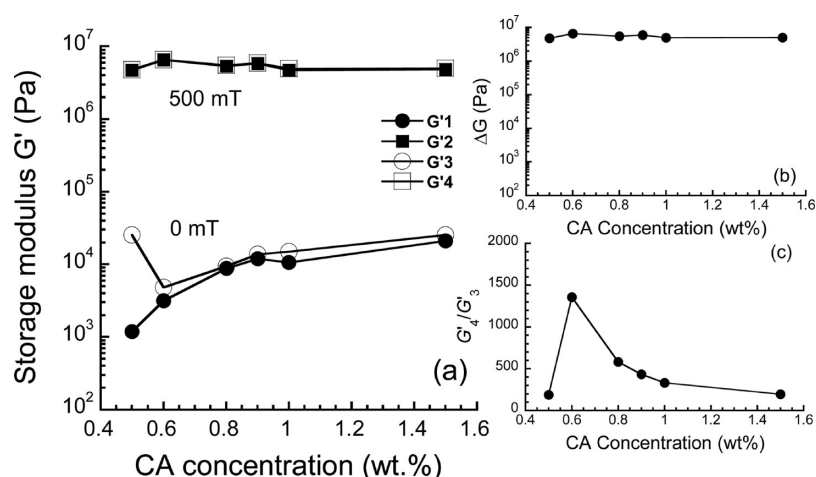
Figure 9a displays the storage modulus in the linear viscoelastic regime as a function of CA concentration when the volume fraction of magnetic particles was constant at 0.30. The storage moduli G'_1 and G'_2 show the modulus at the first application of a magnetic field of 500 mT; the storage moduli G'_3 and G'_4 represent the modulus at the second application of the magnetic field, as seen in Figure 6a. At 0 mT, the G'_1 for magnetic gels above 0.6 wt % was almost the same as that of G'_3, suggesting that the distribution of the magnetic particles is recovered to the original arrangement. When the CA concentration was 0.5 wt %, the storage modulus after applying the magnetic field G'_3 was 2.5×10^4 Pa, which is 20 times

higher than that of G'_1. This indicates that the chain structure remains after removing the magnetic field. The storage modulus at 500 mT (G'_2, G'_4) was approximately 5.4×10^6 Pa, independent of the carrageenan concentration. It can be understood that the magnetic gel exhibits a high storage modulus when the matrix is soft enough to form a chain structure with a perfect alignment of magnetic particles. The mobility of magnetic particles is expected to be lowered when

Figure 9. (a) Storage modulus in the linear viscoelastic regime, (b) absolute change in storage modulus, and (c) relative change in storage modulus for CA/CI magnetic gels as a function of CA concentration.

the magnetic gel has a hard matrix. When the carrageenan concentration was 3.0 wt %, the value of G'_2 was equal to a low value of 1.7×10^6 Pa, indicating that the chain structure is imperfect.

Figure 9b,c shows the absolute change and relative change in the storage modulus as a function of carrageenan concentration, respectively. The absolute change in the storage modulus is that in the reversible regime; that is, $\Delta G' = G'_4 - G'_3$. The absolute change in the storage modulus is independent of the carrageenan concentration because the storage modulus at 500 mT is far higher than that at 0 mT. Figure 9c shows that the relative change in the storage modulus reached a maximum at a carrageenan concentration of 0.6 wt %, at which the maximum value was 1400 with respect to the off-field modulus. The carrageenan concentration is considered to be the critical minimum concentration at which the carrageenan chain is able to recover CI particles to the original location. The maximum value of 1400 is the highest for magnetoelastic soft materials as far as we know. When the carrageenan concentration was 3.0 wt %, the value of G'_4/G'_3 dropped to ~10. This evidence tells us that a low storage modulus at no magnetic field, in other words, "random dispersion of magnetic particles", is needed for magnetoelastic gels, enabling a wide range of modulation of elasticity.

Lastly, shall we consider a reason why the combination of CA and CI makes wide range of modulation of elastic modulus? Of course, the higher magnetizable particle that is used, the larger change in elasticity occurs. Except for the difference in magnetism, a main factor is the particle dispersibility in CA gels, which makes the dynamic range of the elasticity changes widened. When magnetic particles are embedded in gels with high volume fractions, the storage modulus fails to be high because the magnetic particles form a particle network. This causes a high off-field modulus of the magnetic gel. In addition, the particle network is destroyed by magnetic fields, resulting in the reduction of the on-field modulus. On the other hand, magnetic gels with a random dispersion of magnetic particles do not demonstrate both effects that are negative factors for magnetoelasticity. Thus, we consider that the particle dispersibility is a main reason why the CA/CI magnetic gels demonstrate drastic changes in elasticity. Another important factor is adhesion between magnetic particles and the matrix. The magnetorheology of carrageenan gels with iron oxide (CA/

IO gels) was extremely lower than that of CA/CI gels.[26] The relative change in modulus of CA/IO gels is less than 1/10 compared to that of CA/CI gels, although there is no significant difference in both magnetism and particle dispersibility. For example, the saturation magnetization of CI particles (~214 emu/g) is only twice that of IO particles (~94 emu/g). The off-field modulus for the CA/IO gels is nearly equal to that for the CA/CI (~10^4 Pa). We consider that the IO particles were strongly bound within the matrix and the particles cannot form chains effectively under magnetic fields.

4. CONCLUSIONS

The effect of magnetic fields on the storage modulus of magnetic hydrogels consisting of carrageenan and carbonyl iron particles has been investigated by varying rheological parameters, magnetic fields, the volume fraction of magnetic particles, and the carrageenan concentration. The drastic changes in elasticity with an absolute change in the storage modulus of ~4.0 MPa and a relative change in the modulus of 1400 were observed, which is the highest value in the past. The off-field storage modulus of the magnetic gel obeyed the Krieger−Dougherty equation up to a high volume fraction of 0.30, indicating that the particles disperse randomly in the carrageenan gel. At 500 mT, the storage modulus was higher than 4.0 MPa, which is equal to that of magnetic fluids, indicating that the magnetic particles move and form a chain structure by magnetic fields, although the particles were embedded in a cross-linked gel. High dispersibility and high mobility of magnetic particles in the carrageenan gel play a key factor in generating the giant magnetoelastic behavior. Theoretical estimation by Maxwell's stress strongly indicates that the magnetically induced modulus is mainly caused by the stress transfer via the chain structure, not only by magnetic interaction between magnetic dipoles. Morphological studies by scanning electron microscopy revealed the evidence that the magnetic particles embedded in the gel were aligned in the direction of magnetic fields, accompanied by stretching of the gel network. We conclude that the giant magnetoelastic phenomenon observed in carrageenan/carbonyl iron hydrogels originates from the chain structure consisting of magnetic particles, which is similar to magnetic fluids.

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Notes

The authors declare no competing financial interest.

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