

Speckle Analysis of Relaxation Dynamics in Magnetorheological Elastomers

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Introduction

Magnetorheological (MR) elastomers are a new class of engineering materials whose mechanical properties can be controlled by a magnetic field [1]. This is achieved by loading polymeric materials such as natural rubber with a suspension of carbonyl iron particles of size range 0.1 -5 microns [2]. The materials are of interest for automotive applications, specifically for their ability to change their viscoelastic properties under application of a magnetic field.

This is analogous in many ways to the behavior of electro-rheological (ER) fluids and ferrofluidic systems [3]. In particular, the formation of cross-linked chains (see Fig.1) of magnetically coupled particles is thought to be responsible for an enhancement of the elastic shear modulus as the applied magnetic field is increased.

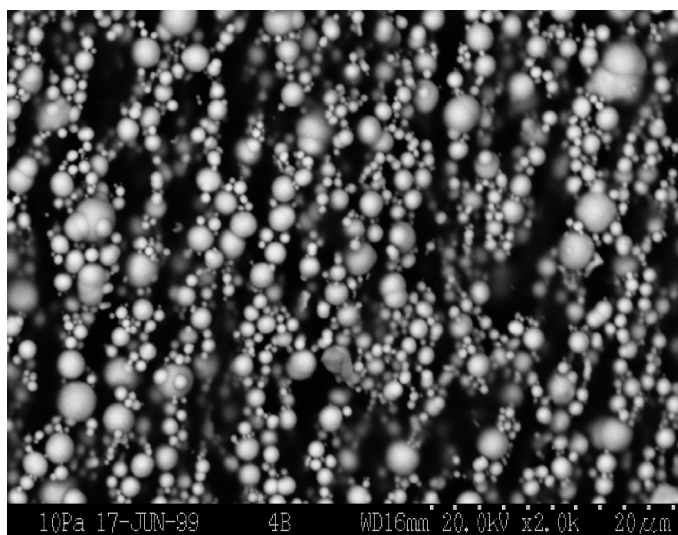


Fig. 1: Scanning electron micrograph showing alignment of iron carbonyl particles in a natural rubber matrix after curing in a magnetic field. 40 μm x 60 μm scan area.

However, there is little information as yet on the structure and dynamics of such chains embedded in a polymer matrix. The purpose of these experiments, which are the first of their kind, is to provide some insight into the relaxation of the MR elastomer when subjected to a magnetic field.

Methods and Materials

X-ray scattering, particularly at small angles, is an ideal tool to study the ordering of magnetic particles embedded in the polymer matrix. The APS offers a special advantage for such studies in the form of *coherent x-rays*. The speckle produced by scattering of transversely coherent x-ray beams can be used to study the relaxation dynamics of the polymer/carbonyl iron network, behavior which is critical for developing applications of this new composite material. We have demonstrated the feasibility of this approach by observing very strong speckle scattering from carbonyl iron particles embedded in natural rubber. In most of the samples studied here, the polymer was cured in a magnetic field of 5-10 kOe in order to impose magnetic order on the embedded carbonyl iron particles. The experiments were performed on MHATT-CAT's undulator beam line, 7-ID, utilizing the pink-beam small-angle scattering facility. The speckle pattern is observed using either a single channel (Amptek) detector, or a direct detection CCD camera. A 20 μm precision slit placed ~ 60 cm after the sample defines the wave vector of the scattered x-rays in the case of single channel data acquisition. The incident beam was collimated by a 5 μm x 5 μm slit with a flux of $\sim 4 \times 10^{10}$ ph/sec. Measurements were recorded at a scattering angle of ~ 5 mR, corresponding to a diffraction vector of 0.02 \AA^{-1} . Although we have not addressed this issue yet, we expect that the dynamics will be wave vector dependent (becoming slower as the x-ray wave vector decreases, extending the length scale of interparticle correlations). Also we expected that the relaxation would be strongly dependent on the magnetic field conditions under which the material is polymerized.

Results and Discussion

Fig. 2 is a time series of the intensity measured with the 20 micron slit/detector combination. The data illustrate the intense speckle scattering that results from the illumination of the MR polymer by transversely coherent x-rays from the undulator source. The x-ray intensity associated with an individual speckle in this sample, largely coming from the strongly scattering carbonyl iron particles, can be as much as several thousand counts per sec.

The intensity variations seen in Fig. 2 are a record of the fluctuating speckle intensity at this particular scattering wave vector. The time dependence of the speckle intensity is a measure of the dynamics of the magnetic particle motion, much in the same way that speckle fluctuations have been used to study the dynamics of colloidal gold clusters [4]. In the case of MR elastomers, the dynamics is presumably controlled by relaxation of the magnetic chain/polymer matrix composite rather than by diffusive motion.

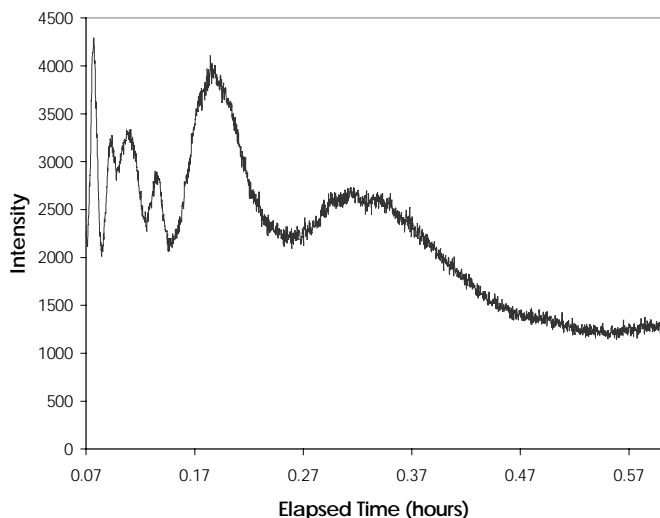


Fig.2: Time series of speckle intensity fluctuations in coherent x-ray scattering from an MR elastomer sample that has been magnetized in a 20kOe field. The horizontal scale is the interval after the sample was removed from the field.

The MR samples are first magnetized in a field of ~ 20 kOe; then the field is turned off and the time series of the speckle intensity is recorded continuously for a period of up to 2 hours. In Fig. 3 it can be seen that the speckle intensity fluctuations are more rapid shortly after magnetization, but become less so with elapsed time. Eventually, the amplitude and frequency of the fluctuations diminishes so that the scattered intensity becomes essentially constant (normalized to the incident beam). In order to quantify this relaxation behavior, we recorded the time intervals between maxima and minima of the intensity and plotted these intervals as a function of elapsed time after magnetization. The resulting curve, consisting of data from five different samples, is shown in Fig. 3. The data fit approximately to an exponential relaxation with a time constant of 0.4 hours. This rather sluggish relaxation is probably associated with the motion of the magnetic chain structure shown in Fig. 1.

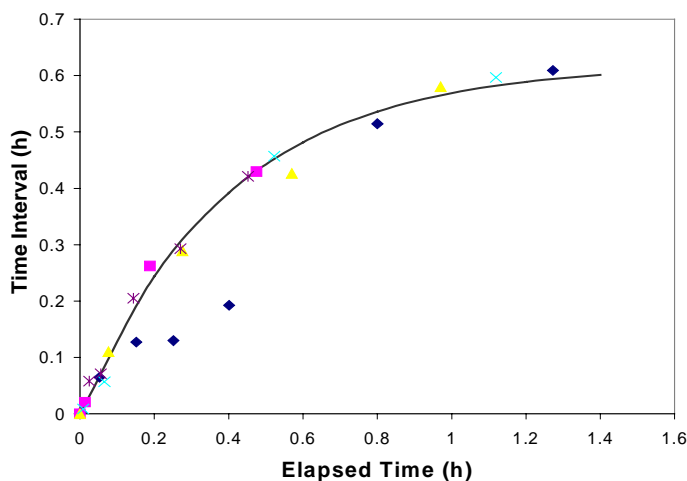


Fig. 3: Exponential relaxation of magnetic particles embedded in natural rubber matrix. The x-axis corresponds to the elapsed time after removing the sample from a magnetic field of 20 kOe. The y-axis refers to the time interval between speckle fluctuations.

It would be interesting to probe the dynamics in real time with time varying magnetic fields and/or applied stress in order to mimic the conditions experienced by the MR elastomer in the intended application of magnetically controlled viscoelasticity. These experiments are ongoing at MHATT-CAT's Sector 7.

Acknowledgments

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