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RAPID COMMUNICATION

Anisotropy properties of magnetic colloidal materials

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Abstract
Magnetic anisotropy of particulate magnetic materials is investigated with respect to the particle shape and compacting geometry. Uniform sized micro-rods and microspheres, coated with nickel, were used. Our results reveal that uniformly aligned rods show the largest magnetic anisotropy, while the field-oriented suspension of coated micro-rods shows somewhat weaker magnetic anisotropy, which is in excellent quantitative agreement with the theoretical calculation. The magnetic anisotropy drops significantly when the micro-rods were replaced by microspheres, even when they were aligned in chain-like formations.

Magnetic composites such as ferrofluids or magnetorheological (MR) fluids are colloidal suspensions of microscopic magnetic particles. It is well known that such magnetic composites possess unique properties when subjected to an external magnetic field. Its viscosity can actually be controlled by the field strength. It is known that when a magnetic field is applied to the colloid, the suspended particles align to form chain-like or columnar structures along the field direction and, thus, dramatically alter the rheological properties of the suspension [1–10]. The magnetic anisotropy of such materials might be useful in applications such as chemical sensors and thermal switches, which has attracted much interest recently [11, 12]. The compacting process of the magnetic particles is believed to play an important role in the anisotropy of magnetic materials [13, 14]. Martin *et al* [15] have studied the relevant contributing factors, including the crystalline magnetic anisotropy as well as the shape anisotropy of the particle in field-structured composites. In this paper, magnetic micro-rods and microspheres, both coated with Ni, are used to investigate the magnetic anisotropy of particulate material with different compacting geometries. We found that the structures formed by the aligned magnetic rods show the largest anisotropy. It is also observed that the magnetic anisotropy becomes weaker when the micro-rods are replaced with microspheres. Moreover, the magnetic property is isotropic for the sample prepared by mixing both magnetic and non-magnetic microspheres randomly. The investigation carried out in this work may provide an easy method for the fabrication of anisotropic magnetic composites.

The magnetic micro-rods and microspheres used in our study consist of 50 μm diameter glass fibres and microspheres onto which a magnetic nickel layer is coated by using the electroless plating technique [16]. In our experiments, the average thickness of the nickel layer for both micro-rod and microsphere is about 8 μm. The SEM image of a section of coated micro-rod can be seen in the upper inset in figure 1(*a*). The samples constructed with rods were made of magnetic rods of lengths 4 and 0.5 mm. Four types of samples were prepared. For sample 1, a regular square grid cell substrate was used to arrange the 4 mm rods along one direction, and then liquid epoxy was added to seal the sample. After the epoxy had hardened, the sample was cut by a diamond saw with dimensions of $3.6 \times 3.6 \times 3.6\,\mathrm{mm}^3$. Samples 2 and 3 were prepared with 0.5 mm micro-rods and microspheres, respectively, and aligned under an external magnetic field and then sealed in epoxy, while sample 4 was made by mixing both magnetic and non-magnetic (uncoated) microspheres uniformly in epoxy under zero field. The hysteresis loops of these samples were measured by VSM at room temperature under an applied magnetic field varying between $-400\,000$ and $+400\,000\,\mathrm{A\,m^{-1}}$. The sample was placed in the central region of a rotatable holder and the data were obtained with different angles between the aligned rod (chain) axis and the field direction.

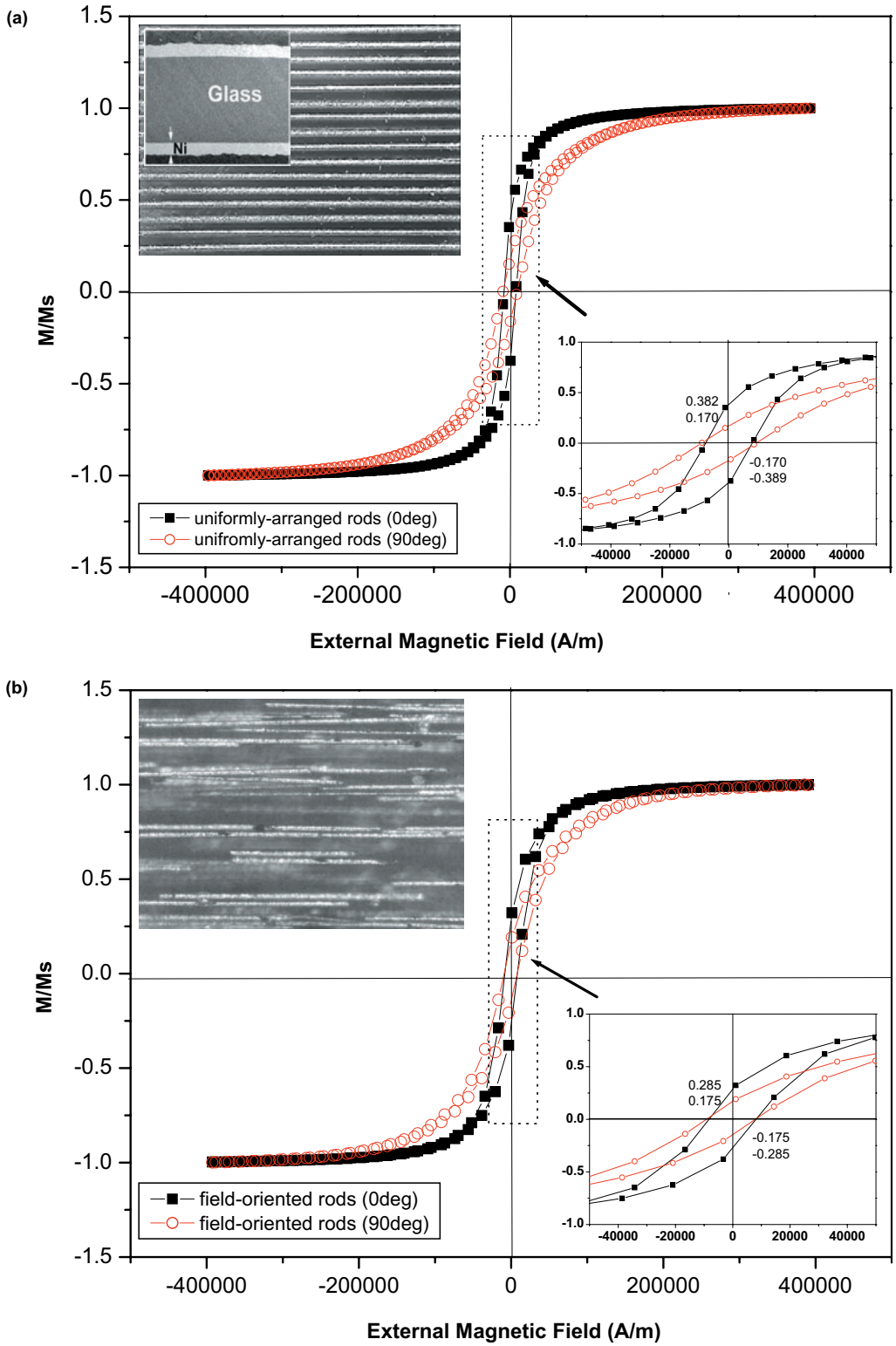

Figure 1. The hysteresis loops measured at $\alpha = 0^{\circ}$ and 90°. Here (*a*) is for sample 1 (uniformly-arranged rods); (*b*) is for sample 2 (field-oriented micro-rods). The upper left insets from (*a*) to (*b*) show the compacting configurations made by different types of particles and the lower right insets are the details of the delineated regions shown by pointed arrows.

The magnetic susceptibility is most commonly defined as $\chi_s = \mathrm{d}M_H/\mathrm{d}H_{\mathrm{ext}}$, where H_{ext} is the applied external magnetic field, and M_H is the component of the magnetic moment along the field direction. It is well known that bulk materials with different physical shapes will have different demagnetization factors; thus, χ_s might be dependent on the actual shape of the samples, which were cut into the shape of a cube for our case (see experimental part for details). In order to eliminate the shape dependence, we may define $\chi = \mathrm{d}M_H/\mathrm{d}H_{\mathrm{in}}$, where H_{in} is the internal field.

For ellipsoidal objects, the external field and the internal field are related as $\vec{H}_{\rm in} = \vec{H}_{\rm ext} - n\vec{M} = \vec{H}_{\rm ext} - n\chi\vec{H}_{\rm in}$. Therefore, $\chi = \chi_{\rm s}/(1 - n\chi_{\rm s})$. Although for a cubic object, the magnetic field inside the material is not a constant, we may also define a demagnetization factor in the sense of averaging over the whole volume. A very accurate expression to describe the dependence of the demagnetization factor of a cube on the susceptibility of the material of which the cube is made is given by $n = 0.274\,40 + 0.147\,35/(\chi + 2.5486)$[15]. Using this we can derive the relation between χ and $\chi_{\rm s}$ for a cubic sample as

$$\chi = \frac{1}{2f}\big((1.147\,35 - 2.5486f) + \sqrt{(1.147\,35 - 2.5486f)^2 + 10.1944f}\big), \quad (1)$$

where $f = f(\chi_{\rm s}) = -0.274\,40 + 1/\chi_{\rm s}$.

In this work, α is defined to be the angle between the applied field and the preferred direction of the micro-rods or chains formed by microspheres. For samples 1, 2 and 3 we measured the magnetic loops starting from $\alpha = 0^\circ$ to 90°, in steps of 15°. In order to measure the magnetic anisotropy, we define the susceptibility anisotropy (relative change in susceptibility) as $\eta = (\chi_{\rm para} - \chi_{\rm orth})/\chi_{\rm orth}$. Here χ is the shape-independent magnetic susceptibility around zero field ($\vec{H} \cong 0$). The subject 'para' indicates the experimental results for $\alpha = 0^\circ$, while 'orth' indicates the experimental results for $\alpha = 90^\circ$.

Using average field theory, the susceptibility anisotropy can be expressed as [15]

$$\eta = \frac{(1/\chi_\perp) - (1/\chi_\parallel) - \psi_z}{(1/\chi_\parallel) - (1/3)(\phi - 2\psi_z)}, \quad (2)$$

where $\chi_\parallel$ is the susceptibility of the constituent particle along the long axis, $\chi_\perp$ the susceptibility of the constituent particle perpendicular to the long axis, ϕ the volume fraction and ψ_z an order parameter. ψ_z is given by the following formula [15]:

$$\psi_z = -\frac{1}{N}\sum_{j=1}^{N}\sum_{i}\left(\frac{a}{r_{ij}}\right)^3 P_2(\cos\theta_{z,ij}). \quad (3)$$

Here, $P_2(x)$ is the Legendre polynomial of order 2, and $\cos\theta_{z,ij}$ is the angle the line of centres between the ith and jth dipoles make to the z-axis. From relation (2) it can be seen that the susceptibility anisotropy for a particulate bulk material can be ascribed to two influencing factors—the shape anisotropy of the constituent particles and the inter-particle interaction which is dependent on the compacting process. It is emphasized here that this calculation is only for cubic packed rods and the average dipolar field is considered.

Comparisons of the measured hysteresis loops for sample 1 (uniformly-arranged rods) and sample 2 (field-oriented micro-rods) are shown in figures 1(a) and (b), respectively. One can observe from figure 1(a) that the susceptibility anisotropy of the sample with uniformly compacted rods is much larger than that of the rods compacted randomly, as shown in figure 1(b). A more detailed comparison is presented in the lower right insets of figures 1(a) and (b), where the anisotropy coefficient is $\eta_1 = 1.52$ for sample 1, and $\eta_2 = 0.97$ for sample 2. The reason for this difference is due to the compacting configuration, since micro-rods were used in both. For sample 1 the coated micro-rods were arranged artificially so that the alignment is perfect. This represents the upper bound to the magnetic anisotropy. For sample 2, the length of micro-rods is shorter than the sample dimension, and they were not perfectly aligned. Many chains aggregated into columns. These imperfections weakened the magnetic anisotropy of sample 2.

Table 1. The theoretical and experimental results for the susceptibility anisotropy for samples 1 and 2.

Configuration	Ψ_z	$\eta_{\rm theory}$	$\eta_{\rm expriment}$
Uniformly-arranged rods	0.26	1.41	1.52
Field-oriented rods			
$n = 3$	0.30	1.09	
$n = 4$	0.35	0.64	0.97
$n = 6$	0.43	0.24	

In order to illustrate the difference between sample 1 (uniformly-arranged rods) and sample 2 (field-oriented micro-rods), using expression (2), we calculated the susceptibility anisotropy η for both cases. For the case of sample 1, 24×24 rods were arranged in a regular square lattice of area of 3.6 mm $\times$ 3.6 mm. For the case of sample 2, we assume that n chains of rods aggregate into one column, and these columns are placed regularly across the cross-section, while the total number of rods remain unchanged. We calculated the values for $n = 3$, 4 and 6. The theoretical calculation is in good agreement with experimental results, and the comparisons are shown in table 1.

The effect of particle shape on the magnetic anisotropy was investigated by replacing the magnetic micro-rods with microspheres. The result is shown in figure 2. It is seen that the anisotropy coefficient η for sample 3 ($\eta_3 = 0.18$) is less than one-fifth of the one for sample 2, i.e. the magnetic anisotropy of sample 3 is much smaller than that of sample 2. Since the compacting process is the same for samples 2 and 3, the difference is attributable to different particle shapes. As seen from the upper left inset of figure 2, the chains formed by magnetic microspheres are similar to those of sample 2. However, in this case, the particles are spherically symmetric, while the coated micro-rods in sample 2 have a much larger shape anisotropy. Therefore, our results show the magnetic anisotropy to be sensitive to the individual particle shape. If the coated and uncoated microspheres were mixed together and randomly compacted, only a small anisotropy coefficient, $\eta_4 = 0.01$, could be detected; i.e. the susceptibility is essentially the same for $\alpha = 0^\circ$ and 90° (see figure 3). This is reasonable, since there is no anisotropy either for the individual particles or for the sample as a whole. For the results shown in figures 1–3, the coercivity is approximately 8000 A m^{-1} for all four samples, in agreement with the results obtained by Martin *et al* [15], which they attributed to the vanishing of the magnetization at the coercive field.

In conclusion, four samples with different particle shapes and compacting configurations were measured to examine the magnetic anisotropy of particulate magnetic materials. Our results reveal the fact that particulate magnetic composites consisting of uniformly arranged rods shows the largest

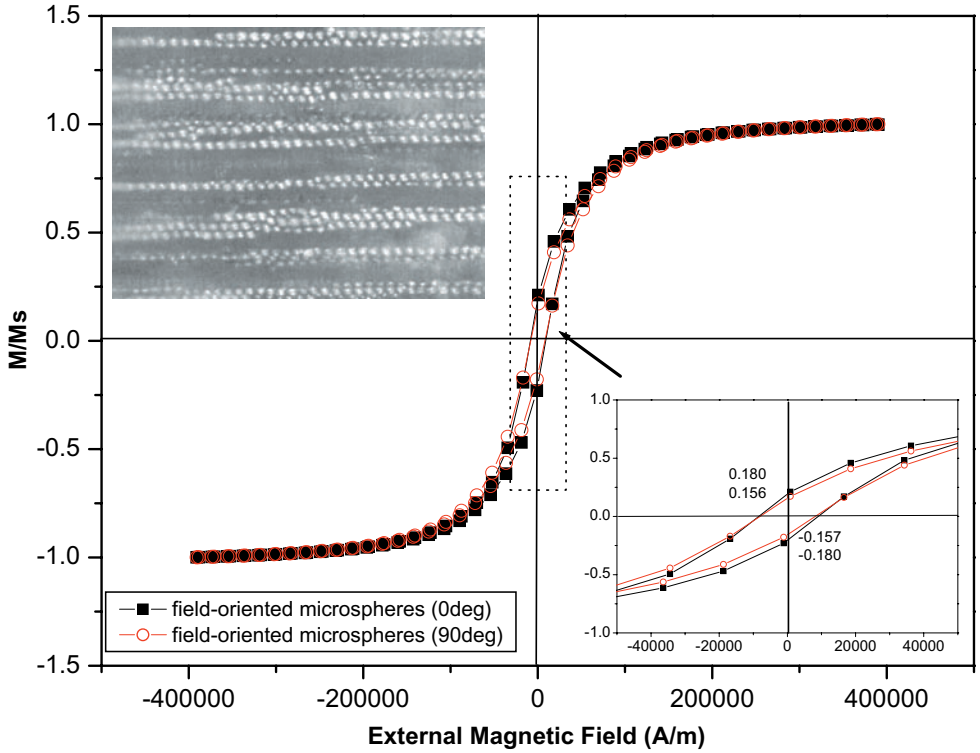

Figure 2. The hysteresis loops measured at $\alpha = 0^{\circ}$ and 90° for sample 3 (field-oriented microspheres).

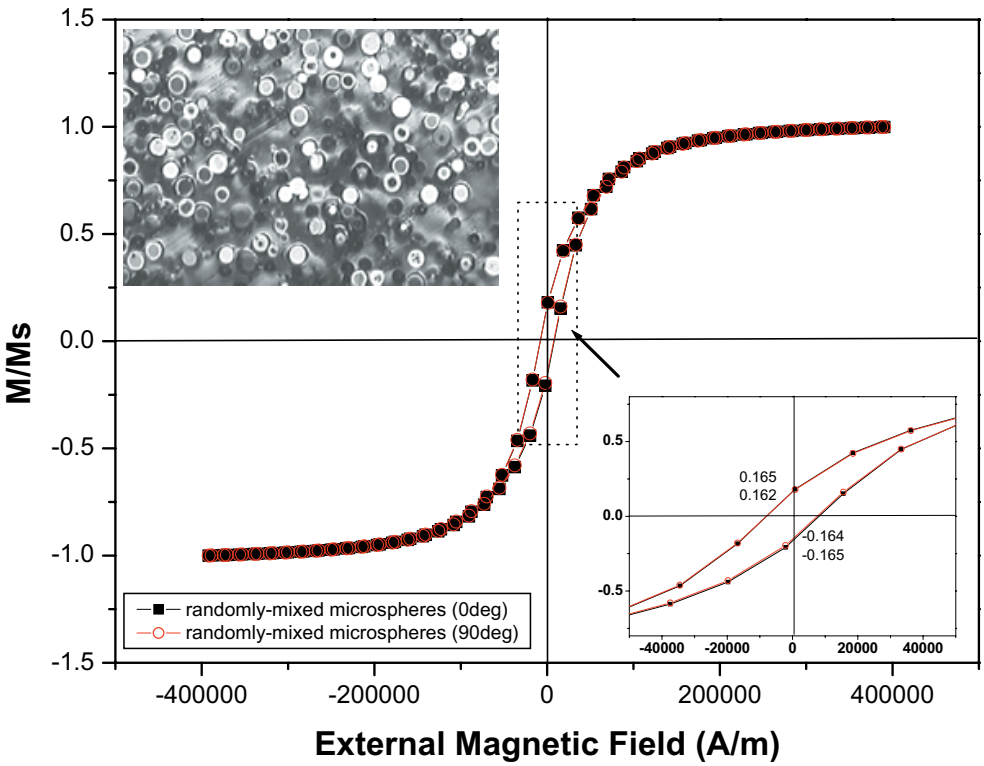

Figure 3. The hysteresis loops measured at $\alpha = 0^{\circ}$ and 90° for sample 4 (randomly-mixed microspheres).

magnetic anisotropy, while the field-oriented chains consisting of micro-rods show somewhat weaker magnetic anisotropy. When the micro-rods were replaced by microspheres, the magnetic anisotropy dropped significantly.

Acknowledgments

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