Effects of Porosity on Coercive Force of Particulate Medium

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It has been suggested that the observed low coercive forces of some magnetic particulate medium, as compared with predictions of single-domain particle theory, may be due to porosity in the particles. Here this possibility is investigated theoretically. When interactions among the pores are considered, the coercive force of a single-domain particle depends on not only the porosity but also the number of pores. In some cases the porosity increases the coercive force, in others it decreases it, while, the magnetization of the particle is always decreasing as porosity increases. The relationship between coercivity and porosity is quite linear. To increase the coercive force, it is concluded that the reduction of porosity is much important than the reduction of the pore number.

I. INTRODUCTION

Consider a single-domain particle in the shape of a prolate ellipsoid. The basic mode of magnetization reversal is coherent rotation of the magnetization.[1] The theoretical predictions for magnetization reversal by coherent rotation have been experimentally verified in cases with predominant magnetocrystalline anisotropy, e.g., chromium dioxide doped with iridium,[2] and cobalteddoped gamma ferric oxide,[3] much lower coercivity and a much broad switching field distribution are obtained than expected for coherent rotation. Instead, the magnetization is reversed in a incoherent fashion by trading long-range magnetostatic field energy against short-range exchange energy. This necessitates that the particle dimensions are large enough to accommodate incremental angles between the spins without introducing too high an exchange energy. Three theoretical models of incoherent rotation modes have been investigated intensively.[4,5] The first mode is buckling, which is a periodic fluctuation of the magnetization direction from the particle axis. This mode involve the magnetostatic field energy and exchange energy. The second mode is curling, which is best described by a bundle of twisted wires along the particle axis. It avoids any magnetostatic energy but involves exchange energy. The third mode is fanning, described by a chain of spheres in which the angle between magnetization and chain axis for neighboring spheres are of opposite sign. This mode relies on magnetostatic energy and excludes exchange energy.

Recently, some self-consistent numerical models were developed to investigate the reversal mechanism of a single particle. A certain new modes were claimed existing, e.g., a flipping mode, which is chain-of-sphere model including the exchange energy.[6] A few studies[7,8] were using the micromagnetics theory to study the nucleation mechanism of an isolated particle, which may not small enough to be a single-domain particle. Simulations using a method which combines the finite element method and the micromagnetics, applied to a perfect ellipsoid were calculated.[9] The nucleation mode is curling and the nucleation field is coincide with the analytic solution. All the methods metioned above, basically, were considering the compromise between the magnetostatic energy and exchange energy.

While the existence of single-domain magnetic particles is now beyond reasonable doubt, some features of their properties are still not understood: notably the inconsistency of the coercivity of some materials, as compared with the theoretical model.[1,4,5,10] Aharoni has indicated that for large particles the preferred mechanism is curling, while for small particles the uniform rotation is preferred.[4] Jacobs and Bean proposed the explanation that elongated particles are equivalent to chains of single-domain spheres.' The specialized models are applicable perhaps in some cases, but certainly not all. Some papers mentioned that the fundamental theory is only valid for idealized particles.[11,12] However, it can be shown that a single-domain particle of any arbitrary shape is precisely equivalent to a suitably chosen ellipsoid with the uniform magnetization.[13] It has been suggested[14,15] that the observed low coercive forces of some magnetic particlate medium, as compared with predictions of single-domain particle theory, may be due to porosity in the particles. For the cases of a single cavity on a single-domain particle was investigated before.[13] The contributions from the interactions among the pores are neglected, while, there are always quite a few pores in the particle. The detailed investigation of that possibility is the purpose of this paper.

In sufficiently small particles, the magnetization is kept approximately uniform by the exchange forces. The critical size, below which this is true, may be smaller for an irregular particle than for the ideal ellipsoid of the Stoner-Wohlfarth theory. We shall assume that for a particle of given shape, a critical size exists, below which the magnetization is to a sufficient approximation uniform, and that the size of the particle being considered is below the critical size for its shape. This is our definition of a "single-domain" particle. In order to fit this approximation the particles must be small enough for the absence of any internal magnetic structure, yet large enough to avoid superparamagnetism. For the magnetic particle with several pores, the exchange interaction is assumed to be strong enough for the magnetization in each region to be uniform.

We first derive the energy expressions, with superposition principle and reciprocity theorem, for an ellipsoidal particle containing several ellipsoidal pores. We neglect crystal anisotropy; in some casess, the equations can be reinterpreted to include it in the manner of Stoner and Wohlfarth.[1]

II. THEORY

We consider the magnetic energy of a porous magnetic particle, as shown in Fig. 1. in a uniform applied magnetic field $\vec{H}_a$. The porous magnetic particle has a magnetization inside

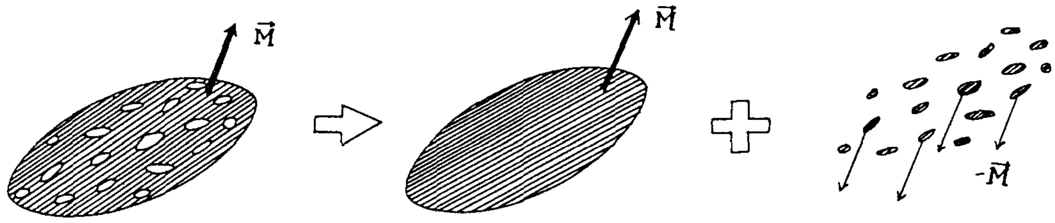

FIG. 1. Decomposition of the magnetization regions of a porous magnetic particle according to the superposition principle.

the volume $V - \Sigma v_i$. The total energy considered is the sum of the Zeeman energy and the magnetic self-energy

$$E_T = E_Z + E_D \tag{1}$$

Invoking the superposition principle (Fig. 1 .), the total demagnetizing field $\vec{H}_D$ can be regarded as the sum of $\vec{H}_g$, from magnetization in volume V, and from magnetization $\vec{H}_p^i$, in each volume v_i. Therefore, the demagnetization energy can be expressed as

$$E_D = -\frac{1}{2}\int_{V-\Sigma v_i} \vec{M} \cdot (\vec{H}_g + \sum_{j=1}^{n} \vec{H}_p^j)dv \tag{2}$$

When interest is confined to uniform magnetization of each ellipsoidal region, the magnetic field $\vec{H}_g$ will be uniform inside each volume V and v_i, but the field $\vec{H}_p^i$ will not be uniform inside the region $V - v_i$. For ease of calculation the magnetic self-energy must be expressed in terms of scaler products of fields and magnetizations both of which are uniform in every region over which a volume integration is performed. This can be accomplished by the reciprocity relationship

$$-\int_V \vec{M} \cdot \vec{H}_p^j dv = -\int_{v_i} (-\vec{M}) \cdot \vec{H}_g dv \tag{3}$$

then from Eq. (2) and (3), we have

$$E_D = -\frac{1}{2}\int_V \vec{M} \cdot \vec{H}_g dv - \sum_{i=1}^{n} \frac{1}{2}\int_{v_i} (-\vec{M}) \cdot \vec{H}_p^i dv$$

$$- \sum_{i=1}^{n} \int_{v_i} (-\vec{M}) \cdot \vec{H}_g dv - \sum_{i,j}{}' \frac{1}{2}\int_{v_i} (-\vec{M}) \cdot \vec{H}_p^j dv \tag{4}$$

For ellipsoidal partical, the demagnetizing field can be expressed as

$$\vec{H}_g = -\vec{N}_g \cdot \vec{M} \tag{5}$$

$$\vec{H}^i_p = -\overleftrightarrow{N}^i_p \cdot \vec{M} \tag{6}$$

Where $\overleftrightarrow{N}$ is the demagnetization tensor.
Therefore, the demagnetization energy can be rewritten as

$$E_D = E_g + \sum_{i=1}^{n} E^i_p + \sum_{i=1}^{n} E^i_{gp} + \sum_{i<j} E^{ij}_{pp} \tag{7}$$

where

$$E_g = \frac{V}{2} \vec{M} \cdot \overleftrightarrow{N}_g \cdot \vec{M} \tag{8a}$$

$$E^i_p = \frac{v_i}{2} (-\vec{M}) \cdot \overleftrightarrow{N}^i_p \cdot (-\vec{M}) \tag{8b}$$

$$E^i_{gp} = v_i(-\vec{M}) \cdot \overleftrightarrow{N}_g \cdot (\vec{M}) \tag{8c}$$

$$E^{ij}_{pp} = \int_{v_i} (-\vec{M}) \cdot \overleftrightarrow{N}^j_p \cdot (-\vec{M})\, dv \tag{8d}$$

In order to calculate the interaction energy among the pores, we need to determine the average interaction energy between two pores. We assume that the center of mass for each pore follows uniform probability to distribute inside the porous magnetic particle. Therfore, the probability of a pair of pores should be correlated as following:

$$\phi_j(\vec{r}_j) = \begin{cases} \frac{1}{V} & r_j \in V \\ 0 & \text{otherwise} \end{cases} \tag{9}$$

$$\phi_i(\vec{r}_i) = \begin{cases} \frac{1}{V-v_j} & r \in V - v_j \\ 0 & \text{otherwise} \end{cases} \tag{10}$$

We also assume the probability of size distribution of pores follow the Gaussian distribution. If we assume that the average size of pores is much smaller than the porous magnetic particle, the average interaction energy for a pair of pores should be

$$<E_{pp}> \cong \frac{1}{V} \left\{ \frac{V_p^2}{n^2} - \frac{\delta^2}{2(n-1)} \right\} \vec{M} \cdot [\overleftrightarrow{N}_g - <\overleftrightarrow{N}_p>] \cdot \vec{M} \tag{11}$$

where δ is the half-width which equals Fv. v is the mean volume of the pore, which equals V_p/n. n is the number of the pore inside the magnetic particle, and V_p is the total volume of pores.

Therefore, the total interaction energy should equal

$$\sum_{i<j} E_{pp}^{ij} = C_2^n < E_{pp} >$$
$$\cong \frac{V}{2} p^2 (\frac{n-1}{n} - \frac{F^2}{4n}) \vec{M} \cdot [\overleftarrow{N}_g - <\overleftarrow{N}_p>] \cdot \vec{M} \tag{12}$$

where p is the porosity, which equals V_p/V. From Eq. (1), we know that the total energy is

$$E_T = -V(1-p)\vec{M} \cdot \vec{H}_a + \frac{V}{2}[1 - 2p + p^2 (\frac{n-1}{n} - \frac{F^2}{4n})] \vec{M} \cdot \overleftarrow{N}_g \cdot \vec{M}$$
$$+ \frac{V}{2}[p - p^2 (\frac{n-1}{n} - \frac{F^2}{4n})] \vec{M} \cdot < \overleftarrow{N}_p > \cdot \vec{M} \tag{13}$$

and the coercive force is

$$H_c = M [\frac{1 - 2p + p^2 (\frac{n-1}{n} - \frac{F^2}{4n})}{1 - P}] (N_g^{\perp} - N_g^{\parallel})$$
$$+ M [\frac{p - p^2 (\frac{n-1}{n} - \frac{F^2}{4n})}{1 - P}] (< N_p^{\perp} > - < N_p^{\parallel} >) \tag{14}$$

III. RESULTS

To simplify, the calculations, we assume that the ellipsoids to be spheroids, the inner spherioids (pores) are indentical in shapes but with various size. If the long axis of magnetic particle and pores coincide, the coercive force can be determined by the Eq. (14). From Fig. 2, the magnetization linearily decreases with increasing porosity. The coercive force

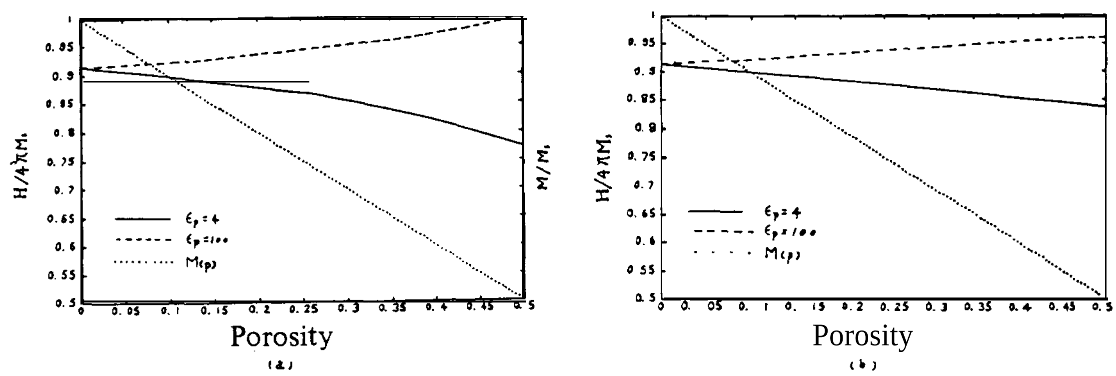

FIG. 2. Porosity dependence of magnetization and coercivity. The dot line is for the magnetization. The solid line is for the coercive force with $\epsilon_p = 4$, while the dash line is for the with $\epsilon_p = 100$. ϵ_p is the acicularity of the inner pores. The acicularity of outer magnetic particle is 8. The half-width is 0.2 V/n and the number of pores, n, in (2-a) is 1, and (2-b) is 10.

can either increase or decrease with increasing porosity. If the acicularity of inner pores is larger than the magnetic paritcle, the coercive force will increase with the existence of the pores. Otherwise, the coercive force will descrese (Fig. 2.). In most cases, the particulate recording medium are acicular particle and the shapes of inner pores are less acicular, therefore, both magnetization and the coercive force will decrease as increasing porosity.

From Fig. 3, we know that the influence of number of pores is only significant at large porosity. The relationship between the coercive force and porosity is quite linear for large

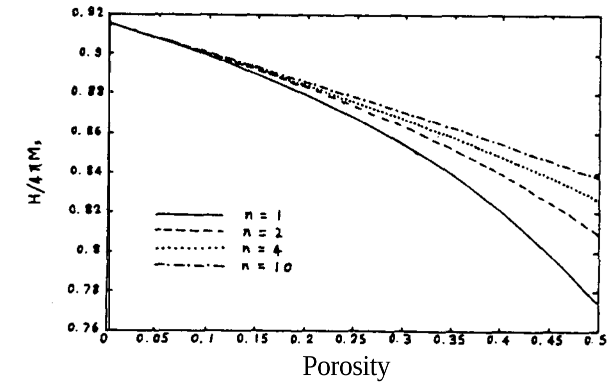

FIG. 3. Porosity dependence of coercivity with different number of pores. The acicularity of outer magnetic particle is **8** and inner pores is **4.** The half-width is **0.2** V/n.

number of pores, while, for the small number of pores, the relationship between the coercive force and porosity is nonlinear. For the same porosity, the coercive force either increases or decreases with increasing number of the pores (Fig. 4.). For the most cases, the acicularity of pores is less than the magnetic particle, therefore, the coercivity increases with increasing number of pores.

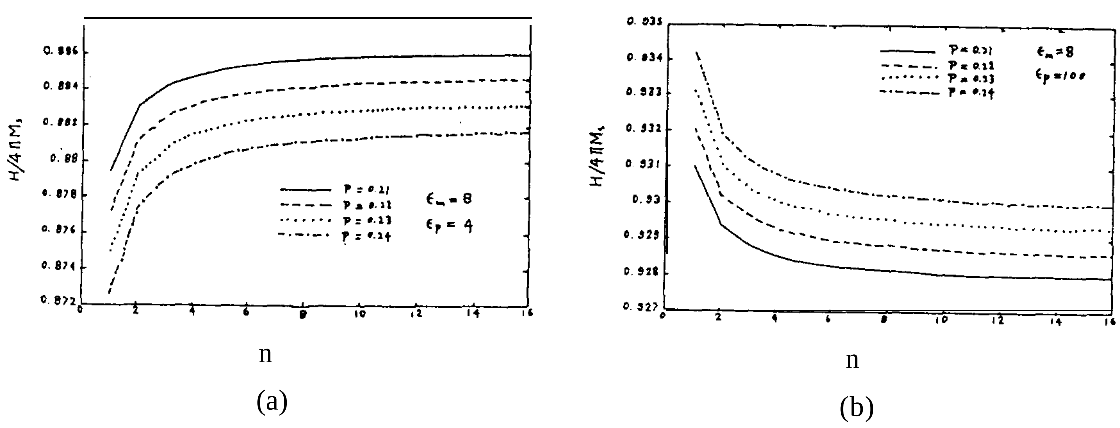

FIG. 4. The relationship between number of pores and coercivity. The acicularity of outer magnetic particle is 8 and the half-width is 0.2 V/n.

In order to investigate the effects of oblique angle for the pores, we calculate the average contribution for a certain polar angle. From Fig. 5, the average contribution equals to zero at a critical angle. If the acicularity of pore is larger, the coercive force will decrease faster with increasing polar angle.

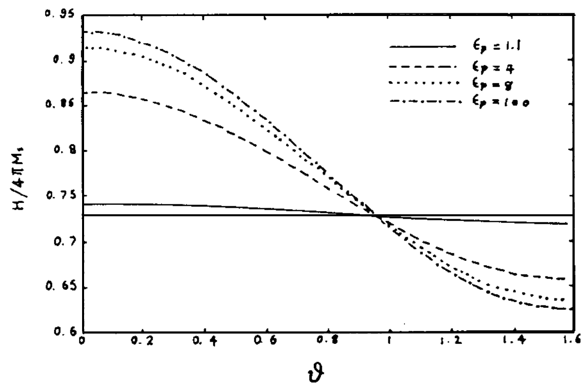

FIG. 5. The relationship between coercivity and polar angle of pores. The acicularity of outer magnetic particle is 8 and pososity is 0.2.

IV. DISCUSSIONS

For the investigation of the dependence of coercive force, we assume that both magnetic particle and pores are ellipsoidal shapes. Although the calculations presented involved ellipsoidally shaped regions of magnetization, the results can be extended to regions of arbitrary shape when the magnetization in each region is uniform.[13] The magnetic properties of magnetic particle with several pores are summarized as follows: (1) The magnetization per unit volume decreases with increasing porosity. (2) The pores can either increase or decrease the coercive force. It is determined by the relative shapes between the pores and the magnetic particle. (3) Unless the number of the pores is very small, the coercive force linearily depends on the porosity. (4) The influence of number of pores on the coercive force is small. Therefore, to increase the coercive force, it is concluded that the reduction of porosity is much important than the reduction of the pore number. (5) The coercivity is insensitive to the pore size variation. (6) If the polar angle of pore is not colinear with easy axis of magnetic particle, the nucleation will begin at lower applied field.

It has been demonstrated that several ellipsoidal cavities in a ellipsoidal single-domain particle can either increase or decrease the coercive force; but if the coercive force without the cavities is large, the usual effect of the cavities, and in some cases the only possible effect, is a decrease. This is therfore a mechanism capable of contributing to the observed coercive force of some particulate recording medium.

We can also use the similar method to consider the mean effect of interactions on the coercive force of an assembly of magnetic single-domain acicular particles. The magnetic interaction energy can be calculated and related to the coercive force. The coercivity is first degree equation in packing density,[16] which was verified with the experimental data.[17]

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