The microstructure and computed magnetic properties in nanoscaled permanent magnets

J.X.Zhang_T.J.Zhang_K.Cui

(Key State Laboratory of Die Technology, HUST ,Wuhan ,China)

ABSTRACT

Magnetic properties of a model nanocomposite magnet were calculated by means of computer simulation, and the effects of the amount of the soft magnetic phase on them were studied. The numerical results show that the remanence enhanced with the increase of the soft grains, it is due to the high spontaneous magnetication of soft magnetic phases and exchange interactions. The nonuniform grains have no evident effect on the remanence of the magnet, but the coercivity decreased.

Keywords: nanocomposite magnet; computer simulation; remanence enhancement; coercivity; irregular model

1. Introduction

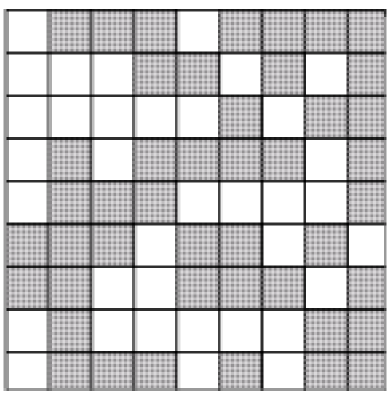
A nanocomposite structure comprising the highly anisotropic $Nd_2Fe_{14}B$ phase and soft-magnetic a-Fe phase has opened a way for a new generation of permanent magnets with enhanced remanence Br and energy products $(BH)_{max}$. The high performance of these magnets arises from the exchange coupling between the soft- and hard-magnetic phases. It must be noted, that the magnet properties of the nanocomposite magnets are not the intrinsic property, which means they are depend not only on the chemical composition, the temperature and the magnetic anisotropy but also strongly on the microstructure of the materials. Now one of the focus of nanocomposite magnets research is how to control the exchange coupling, by modify the microstructure of the magnet, as a consequence the improved squareness, higher remanence and the not severely reduced coercity bring about an enhancement of $(BH)_{max}$. It is important therefore to develop quantitative models that demonstrate the influence of grain-grain coupling, grain size and grain size distribution on the behaviour of the material, models that will enable us to better understand the behaviour of existing materials and suggest ways in which they may be improved.

Several micromagnetic models of these polycrystalline magnets that take into account both dipolar and exchange interactions between grains have been treated in the literature. In one very economical approach, that of Fukunaga and Inoue[2][3], the representation of the magnetic structure of each grain is restricted to a single computational element. The magnetization within each grain is treated as uniform, of magnitude Ms but of variable orientation. The exchange interaction between nearest-neighbor grains is represented by an energy _$J_e S \vec{m}_i.\vec{m}_j$ where J_e is an exchange constant, $\vec{m}_i, \vec{m}_j$ are unit vectors parallel to $\vec{M}_i, \vec{M}_j$,the magnetization vectors of the two grains, and S is their area of contact. The outstanding advantage of the Fukunaga's model lies in its great computational economy, enabling statistically representative assemblies of numberous grains to be treated with full inclusion of their dipolar interactions. Their comparison of hysteresis loops calculated both with and without including dipolar interactions in the model, demonstrates convincingly that, in such a highly anisotropic material as $Nd_2Fe_{14}B$, the dipolar interactions, that are so demanding of computational effort, may be justifiably be ignored. But in the model all domain walls are necessarily confined to the grain boundary. Any possibility of incoherent magnetization changes within the grains. e.g. fanning ,curling, buckling and domain wall motion, is excluded a priori. However, in reality ,such processes provide the dominant mechanisms for magnetization reversal in grains somewhat larger than the domain wall thickness $\_{W,}$ their neglect must be expected to lead to a serious overestimate of the coercivity.

By contrast in their treatment of the problem, Fischer et al have adopted an essentially orthodox micromagnetic approach. These authors have treated two-dimensional an three-dimensional models, employing computational meshes finer than the grain size to represent the magnetization distribution of the assembly, and using finite element approximations implement numerically the standard equations of micromagnetics that relate to the exchange energy, anisotropy energy and Zeeman energy of interaction with the applied field. Their three-dimensional models usually consist of a cube divided into 35(occasionally 64) $Nd_2Fe_{14}B$ grains, of various sizes, in either a regular or an irregular arrangement, The easy axes of grains were oriented at random. Since each grain is subdivided into 200-600 tetrahedrons, the model account for non-uniform magnetization distributions arising from strayfield or exchange effects within each grain. This is a great effort compared to previous approaches by Fukunaga. But the increase of the number of magnetization vectors from 1 to 200-600 per grain leads to an enormous increase of the computation time. Therefore, the maximum number of grains is limited. The statistical variations between different computational samples with small numbers of grains lead to wide variations in their magnetic properties.

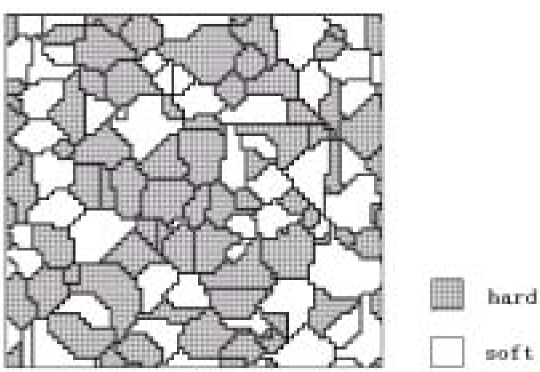
It is evident that further computational studies are needed to establish a more satisfactory match between experiment and realistic models based on micromagnetic theory. In this work, we modify the model of Fukunaga, subdivided the grains to incorporate nonuniform magnetization reversals while maintaining its advantage of computational economy. We also build grain structures to study the effect of irregular grains on magnets properties. Instead of the FEM and FDM general used, a new way simulating the spins rotate has been used, to calculated the magnetization process. It should be noted that, it need 100-200h to get a computational result in Fischer's research, but it only need 20-30h in our work (in common PC), so that we can include more grains in our model. It opens a way for computer simulation of nanoscaled permanent magnets.

2.Simulation model and method



(a)

2.1 Microstructures of the models



(b)

Fig 1. The section of regular grains model(a) and irregular grains model(b)(40% soft phase)

Our regular simulation model is composed of 9_9_9 cube-shape grains. In order to study the effects of nonuniform magnetization reversals ,all the grains were subdivided in 8^3 cubic element_assuming an uniform magnetization distribution within each element. To investigate numerically the influence of the irregular grains, a model simulating the grains growth has been use: In the given model, firstly subdivided it into 72^3 cubic elements. Then starting from the randomly located seed points (9^3), grains grow with constant growth velocity in each direction. This modeling of grains yields realistic three-dimensioned microstructures. The section of the regular and the irregular grains model shown in Fig 1. The easy axis of each grain was selected at random and all elements composing any given grain have parallel axes.

1.2 The calculation of Gibbs free energy

In the continuum theory of micromagnetics, if the magnetoelastic and surface anisotropy effects are neglected, the magnetic Gibbs free energy $\_{t}$ of a ferromagnetic specimen in an applied magnetic field is the sum of several energy terms _

$$\_{t}=\_{ex}+\_{k}+\_{H}+\_{str} \quad (1)$$

Where $\_{ex}$ is the exchange energy_$_{k}$ is the magnetocrystalline anisotropy energy_$_{H}$ is the magetostatic energy_$_{str}$ is the stray field energy_Several numerical results show ,the dipolar interaction may be ignored safely, so in this work, the magnetic Gibbs free energy can be expressed as _

$$\_{t}=\_{k}+\_{ex}+\_{H} \quad (2)$$

For every element ,the total energy is given by_

$$\Phi_{ti}=K_iV(\sin^2\theta_i\ )+\frac{J_eS}{6}(\sum_{j=1}^{6}\vec{m}_i.\vec{m}_j)+J_{si}V(\vec{m}_i.\vec{H}) \quad (3)$$

For the magnet

$$\Phi_t=\sum_{i=1}^{N}\{K_iV(\sin^2\theta_i)+\frac{J_eS}{6}(\sum_{j=1}^{6}\vec{m}_i.\vec{m}_j)+J_{si}V(\vec{m}_i.\vec{H})\} \quad (4)$$

where_N is the number of the elements in the magnets_72^3_. K_i, J_{si} are the anisotropy constant and saturation magnetization, and their values depend on the kind of grain in which the element is located .$\_{i}$ demotes the angle between magnetization and the easy direction in the ith element. S,V are the surface area and the volume of each element_J_e is the exchange interaction constant per unit surface area, and a single value was assumed for all the boundaries. $\vec{m}_i(\vec{m}_j)$ are the magnetizations in the ith and jth element. For the caculations the material parameters of $Nd_2Fe_{14}B$ [5],_K_1=4.3_10^6J/m^3,K_2=0.65_10^6J/m^3,J_s=1.63T_, and _-Fe (K_1=4.6_10^4J/m^3,K_2=1.5_10^4J/m^3,J_s=2.15T,Temperature is 300K) have been used.

1.3 The caculation of demagnetization process

As only quasistatic magnetic states of the system are being considered here, the distribution of magnetization in the model was assumed to be such as to constitute a stable, or metastable equilibrium state, such that the magnetic energy E of the cubic computational region is at a minimum. Instead of the FEM and FDM general used, a new way simulating the spins rotate has been used, to calculated the magnetization process

In the real process of magnetization the spin of every grain rotated simultaneously in the external field. But in this work, for the economy of computation, in the given external field, we rotate the spin of only one element to get the stable state. Repeat the work for every element, then scan the whole magnet. After every scan, sum the free energy of the whole magnet if the difference between that of the nth and (n+1)th fulfill the control precision, stop the scanning, then we assume the system get the stable state, else continue, till the condition can be met. Then calculate the average of the magnet polaration and write down the external field.

2_Results and Discussion

2.1 The effects of soft phase.

2.1.1 Remanence enhancement

According to the Stoner-Wohlfarth theory, which assumes non-interacting homogeneously magnetized single-domain particles $J_r / J_s = 1/2$ .The remanence for an assembly of non-interacting $Nd_2Fe_{14}B$ (randomly oriented)and _-Fe is given by (dashed line in Fig.5(a))_

$$J_r \approx 0.81T + \frac{V^{soft}}{V^{total}} \times 0.27T \qquad (5)$$

where V^{soft}/V^{total} denote the volume fraction of hard magnetic grains and soft magnetic grains, respectively.

The remanence increases linearly with the amount of_-Fe_but there is still a great discrepancy between the dashed line and the numerical result in Fig 2. The remanence enhancement, according to isolated hard and soft magnetic grains, has to be entirely attributed to interpartical exchange interactions. For explanation, we can briefly consider the microscopic behaviour of underlying model magnet by going from the saturation state to the remanent state. In micromagnetism, the stable equilibrium state of Js can be obtained by minimizing the total energy. According to Equ(1) the total energy is the sum of several energy terms, which can be characterized by their effects on the magnetic polarization Js. The magnetostatic energy $\_{H}$ tries to rotate Js parallel to the applied field. The magnetocrystalline anisotropy energy $\_{k}$ causes Js to be preferably oriented along certain easy axes. The exchange energy $\_{ex}$ aligns the magnetic moments parallel.

For high external fields, the magnetostatic energy determines the distribution of J_s_r_, so that all spins rotate parallel to the applied field . Therefore $\_{k}$ is very high and $\_{ex}$ has almost vanished . As the external field drops, $\_{k}$ decreases because the magnetic polarization of each individual grain rotates towards its particular easy axes. Therefore strongly inhomogeneous distributions of $J_s(r)$ arise at the boundaries between misoriented grains. However the competitive effects of decreasing magnetocrystalline anisotropy and increasing exchange interactions cause a smooth transition of Js from one easy axis direction to the other. As a consequence, the resultant polarization to the field direction is increased compared with the remanence of non-interaction particle.

2.1.2 Coercivity

Up to now, we considered the behaviour of two-phase permanent magnets only from saturation state to remanent state. With increasing opposite external field, the spontaneous magnetic polarization starts to deviate from the easy axes within the grains. Coming up to the coercitive field, nearly all magnetic moments reverse their directions. Since the low anisotropy of soft magnetic phase, its spins reverse at low external field. It resulted in the disaccord of spin arrangements in grain boundary between hard magnetic phase and soft magnetic phase. The state is unstable, finally cause magnetization reversal in neighboring hard magnetic phase and thus determine the coercive field. When the amount of_-Fe is small, the exchange effect between it and neighboring hard magnetic phase restrain its too earlier reversal, so that keep the

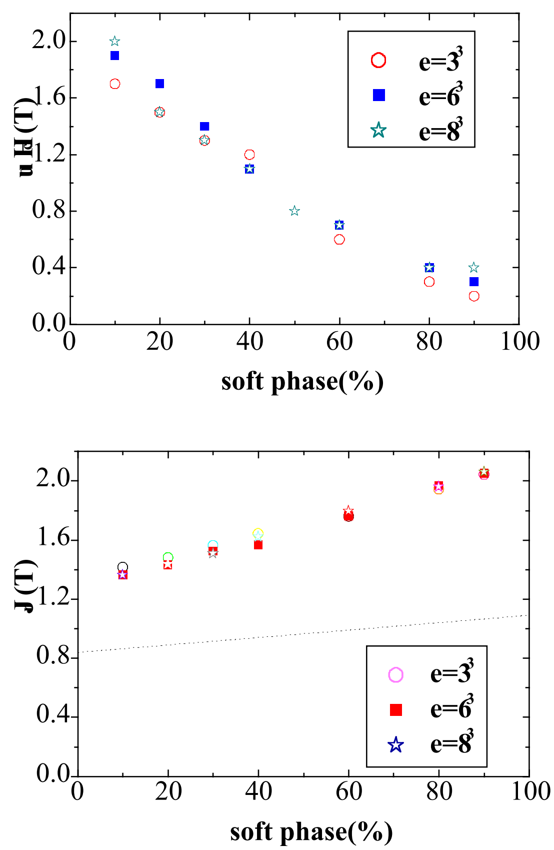


Fig 2. Remanence (a) and Coercivity(b) as a function of the amount of soft magnetic phase. (with different number of element e) Within the framework of the Stoner-Wohlfarth theory, the remanence of nanocomposite magnet_assumes non-interacting single-domain particles_is given by the dashed line.

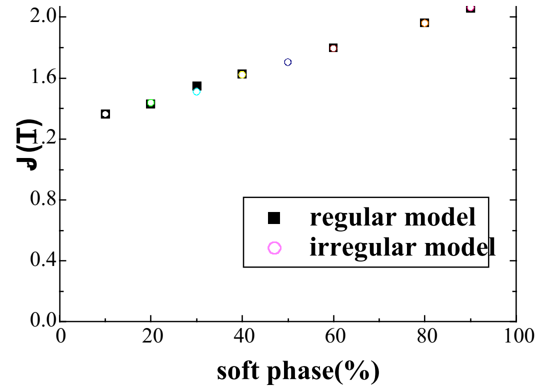
coervity in a considerable level. But the extension of connected soft magnetic regions increases with increasing percentage of_-Fe, the effect of exchange is unimportant then. Therefore the coercive field decreases with increasing amount of_-Fe, as we can see in Fig 2.

2.1.3 The divided elements of every grain

In our model_we made improvement of Fukunaga's model. Every grain was divided into many elements to study the nonuniform magnetization in the grains. Generally the nonuniform magnetization results in the reseral, so that the division of the grains should be cause the low coervity in the results. The results of Griffiths M K et al validate it. But in our work _the number of element that every grains divided has no evident effect on the coercity, the mechanism need further research. The point need to be paid attention to is that the result of Kuma is according to ours, and the exchange effect is also represented as $-J_e S\bar{m}_i.\bar{m}_j$ in his research, but in Griffiths M K' work there in no such approximation. This may be the reason for the difference between the two research results.

2.2 irregular grains

The numerical results, presented Fig.3, allow us to compare the magnetic properties of two-phase permanent magnets with regular and irregular shaped grains. While there is no drastic different between remanence, the irregular shape has a profound effect on the coercivity, they were according to these of Fischer's. As Fischer's explanation[5], the difference is due to the inhomogeneous distributions of the spontaneous polarization near the grain boundiaries which came from the sharp corners and edges of the irregularly shaped particle. The resulting local magnetic volume charges yield strong inhomogeneous demagnetizing fields, which generally reduce the switching fields of particles. But we do not take into account the dipolar interaction, so the discussion of Fischer isn't complete. For the irregular grains, the coercivity loss came from the big grains, for which the exchange effect between it and neighboring hard magnetic grain can not constrain its reversal effectively. For more soft magnetic phase, the difference between regular and irregular model vanishes because the extension of connected soft magnetic regions increases, the effect of irregular grains could be ignored safely. The few hard magnetic grains are completely surrounded by soft magnetic grains. Therefore, the soft phase begins to govern the properties of the two-phase magnet.



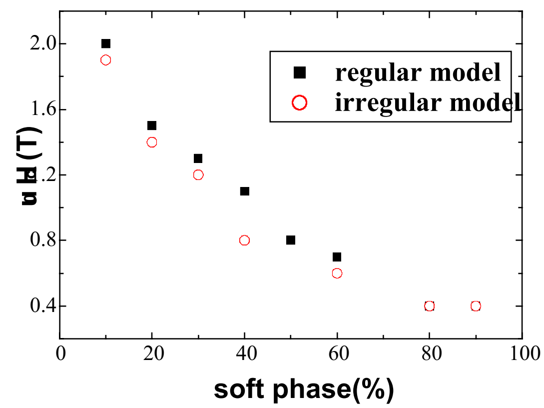


Fig 3. Remanence and Coercivity as a function of the amount of soft magnetic phase. (irregular model/ regular model.)

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