A Simple Analytical Model for Magnetization and Coercivity of Hard/Soft Nanocomposite Magnets

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We present a simple analytical model to estimate the magnetization (σs) and intrinsic coercivity (Hci) of a hard/soft nanocomposite magnet using the mass fraction. Previously proposed models are based on the volume fraction of the hard phase of the composite. However, it is difficult to measure the volume of the hard or soft phase material of a composite. We synthesized Sm2Co7/Fe-Co, MnAl/Fe-Co, MnBi/Fe-Co, and BaFe12O19/BaM/Fe-Co composites for characterization of their σs and Hci. The experimental results are in good agreement with the present model. Therefore, this analytical model can be extended to predict the maximum energy product (BH)max of hard/soft composite.

There are two issues in rare-earth (RE) permanent magnets (PM) for full applications. One is RE mineral security, and the other is a low Curie temperature of Nd-Fe-B magnet. The figure of merit of PM is its maximum energy product, (BH)max. The (BH)max can be estimated as (BH)max = (Br)2/4 for Hci > Br/2 or (BH)max = (Br − Hci)Hci for Hci < Br/2. Br is the remanent magnetic flux density, and Hci is the intrinsic coercivity, which is mainly controlled by the magnetocrystalline anisotropy constant (K). Therefore, high Br and Hci are needed for a large (BH)max. In addition, the PM must also have a corresponding high Curie temperature (Tc) to retain the figure of merit at typical operating temperatures. In an effort to increase the (BH)max of RE-free permanent magnets, concepts of exchange coupling between hard and soft magnetic phases have been proposed. Exchange coupling makes full use of high Hci from the hard phase and Br from the soft phase of a hard/soft composite magnet. Therefore, a large (BH)max of a composite magnet can be achieved. In the magnetic exchange coupled composite, the magnetization direction of the soft phase is pinned to the magnetization direction of the hard phase. This implies that the exchange coupled two-phase magnet behaves like a single-phase magnet. However, the soft magnetic phase needs to be thinner than twice the domain wall thickness (2δw) of hard magnetic phase for full exchange coupling. Thus, the increasing rate of (BH)max with the amount of soft phase is limited. Although the previously proposed models predict the magnetization in the unit of emu/cm³ (M) and K of an exchange coupled thin film magnet reasonably well, a model directly applicable to a powdered (bulk) hard/soft nanocomposite magnets is not yet reported. In this paper, we developed a model for the magnetization in the unit of emu/g (σs) and Hci of powdered hard/soft composite based on, experimentally accessible, the mass fraction of hard and soft magnetic phases instead the volume fraction. The prediction of the developed model was compared with the experimental σs and Hci of Sm2Co7/Fe-Co, MnAl/Fe-Co, MnBi/Fe-Co, and BaFe12O19(BaM)/Fe-Co, where Sm2Co7, MnAl, MnBi, and BaM are hard magnetic phases, and Fe-Co is a soft magnetic phase.

Derivation of Equations

We now derive the equations for σs and Hci in terms of mass fraction of composite. According to theoretical studies on a two-phase composite magnet, the saturation magnetization and anisotropy constant of a composite can be expressed as:

\[ M = f_h M_h + f_s M_s \]  

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and

\[ K = f_s K_s + f_h K_h, \]  

(2)

where \( M \) is the saturation magnetization, \( K \) is the magnetocrystalline anisotropy constant, and \( f \) is the volume fraction. \( h \) and \( s \) in the subscript denote hard and soft phases, respectively. Because of the experimental difficulty of obtaining \( M \) and \( K \) (per unit volume) of powdered composite, we seek to develop expressions for \( \sigma_s \) and \( H_{ci} \) (per unit mass) of a two-phase magnetic composite using experimentally accessible \( \sigma_s \) and \( H_{ci} \) for both hard and soft phases. Noting that \( M \) in Eq. (1) is magnetic moment per unit volume (typically in the unit of emu/cm\(^3\)), they can be expressed as:

\[ M = \frac{\text{magnetic moment}}{\text{volume}} = \frac{\text{magnetic moment}}{\text{mass}} \cdot \frac{\text{mass}}{\text{volume}} = \sigma \cdot \rho \]

where \( \sigma \) is the saturation magnetization (per unit mass in the unit of emu/g) and \( \rho \) is the mass density (in the unit of g/cm\(^3\)). Therefore, the \( \sigma \) (the subscript \( s \) will be omitted for now to avoid the confusion with quantities for soft phase) of two-phase magnetic composites can be written as:

\[ \sigma = \frac{\sigma_s \rho_s f_s + \sigma_s \rho_s f_h}{\rho_s f_s + \rho_s f_h}. \]  

(3)

\( H_{ci} \) due to magnetocrystalline anisotropy\(^5\) is

\[ H_{ci} = \frac{\alpha K}{\sigma \rho}, \]  

(4)

where \( \alpha \) is a constant dependent on the crystal structure and degree of alignment. \( \alpha = 2 \) in the case of aligned particles\(^8\) while for unaligned (random) particles, \( \alpha \) can have different values for different crystals (for instance, 0.64 for cubic crystals\(^7\) and 0.96 for uniaxial crystals). Then, \( H_{ci} \) of the two-phase magnetic composite can be modified to equation (5) by combining Eqs (2) and (4):

\[ H_{ci} = \frac{\alpha (1 - f_s) K_s + f_s K_h}{(1 - f_h) \sigma_h + f_h \sigma_s}. \]  

(5)

By replacing \( K \) in Eq. (5) using Eq. (4), Eq. (5) becomes

\[ H_{ci} = \frac{\sigma_s V_h \rho_s f_s + \sigma_s V_h \rho_s f_h}{\sigma_s V_h \rho_s f_s + \sigma_s V_h \rho_s f_h}. \]  

(6)

where \( H_h \) and \( H_s \) are the intrinsic coercivities of hard and soft phases, respectively. Therefore, the \( H_{ci} \) of a composite can now be estimated by experimental \( H_h \) and \( H_s \) instead of the \( K_h \) and \( K_s \). Furthermore, since it is difficult to measure the volume fraction of a powdered sample, we further develop equations for \( \sigma \) and \( H_{ci} \) of a two-phase composite in terms of the mass fraction. Since \( f_h \) is the volume fraction of hard magnetic phase, i.e.,

\[ f_h = \frac{V_h}{V_h + V_s}, \]  

(7)

where \( V \) is the volume. Therefore, Eqs (3) and (6) become

\[ \sigma = \frac{\sigma_s V_h \rho_s f_s + \sigma_s V_h \rho_s f_h}{V_h \rho_s f_s + V_h \rho_s f_h}. \]  

(8)

and

\[ H_{ci} = \frac{\sigma_s V_h \rho_h f_h + \sigma_s V_h \rho_s f_s}{V_h \rho_h f_h + V_h \rho_s f_s}. \]  

(9)

respectively. Dividing both the numerator and denominator in Eqs (8) and (9) by the total mass, i.e. \( (V_h \rho_h + V_s \rho_s) \), we get:

\[ \sigma = \frac{\sigma_s V_h \rho_h + \sigma_s V_h \rho_s}{V_h \rho_h + V_h \rho_s}, \]  

(10)

and

\[ H_{ci} = \frac{\sigma_s V_h \rho_h + \sigma_s V_h \rho_s}{V_h \rho_h + V_h \rho_s}. \]  

(11)
The mass fraction of hard \((f^m_h)\) and soft \((f^m_s)\) magnetic phases are

\[
f^m_h = \frac{V_h \rho_h}{V_h \rho_h + V_s \rho_s} \quad \text{and} \quad f^m_s = \frac{V_s \rho_s}{V_h \rho_h + V_s \rho_s},
\]

respectively, where \(V_h \rho_h\) is the mass of hard phase and \(V_s \rho_s\) is the mass of soft phase. Accordingly, Eqs (10) and (11) become

\[
\sigma = \sigma_h f^m_h + \sigma_s (1 - f^m_h)
\]

and

\[
H_i = \frac{\sigma_i H_i f^m_h + \sigma_s H_i (1 - f^m_h)}{\sigma_h f^m_h + \sigma_s (1 - f^m_h)},
\]

respectively.

Eqs (13) and (14) can now be used to estimate the \(\sigma_i\) and \(H_i\) of a two-phase magnet by only considering the mass fraction \((f^m_h\) or \(f^m_s)\) of hard and soft phases if their saturation magnetization and intrinsic coercivity are known.

**Experimental Validation**

In order to validate the efficacy of Eqs (13) and (14), we synthesized four different composites, \(\text{Sm}_2\text{Co}_7/\text{Fe-Co}\), \(\text{MnAl}/\text{Fe-Co}\), \(\text{MnBi}/\text{Fe-Co}\), and \(\text{BaM}/\text{Fe-Co}\), by mixing hard and soft magnetic particles in an appropriate weight ratio and characterized them for magnetization and coercivity. It was noted that three different Fe-Co compositions, i.e., \(\text{Fe}_{50}\text{Co}_{50}\), \(\text{Fe}_{65}\text{Co}_{35}\), and \(\text{Fe}_{80}\text{Co}_{20}\), were used for \(\text{Sm}_2\text{Co}_7/\text{Fe-Co}\) composites. The \(\sigma_i\) and \(H_i\) of \(\text{Fe}_{50}\text{Co}_{50}\), \(\text{Fe}_{65}\text{Co}_{35}\), and \(\text{Fe}_{80}\text{Co}_{20}\) are 236 emu/g and 75 Oe, 240 emu/g and 80 Oe, and 232 emu/g and 65 Oe, respectively.

Figure 1(a) and (b) show the \(f^m_h\) dependence of \(\sigma_i\) and \(H_i\) for \(\text{Sm}_2\text{Co}_7/\text{Fe-Co}\) composite with various compositions of Fe-Co. The \(\sigma_i\) decreases linearly as the amount of hard phase \((\text{Sm}_2\text{Co}_7)\) increases in Fig. 1(a). The experimental results (open symbol) are well fitted to our developed equation (13) (solid line). It is noted that at lower concentration of hard phase, deviation of experimental \(\sigma_i\) from the solid (theoretical) line is getting larger.

In Fig. 1(b), experimental \(H_i\) is excellently fitted to the developed equation (14), especially, for the composite with \(\text{Fe}_{65}\text{Co}_{35}\).

As shown in Fig. 2(a) and (b), the \(\sigma_i\) of \(\text{MnAl}/\text{Fe-Co}\) composite linearly decreases by increasing the content of hard phase, and the \(H_i\) increases by following the developed equation (14). Both experimental \(\sigma_i\) and \(H_i\) are in good agreement with the present model.

As shown in Fig. 3(a) and (b), the \(\sigma_i\) of \(\text{MnBi}/\text{Fe-Co}\) composite decreases by increasing the content of hard phase, and the \(H_i\) increases by following the developed equation (14). Both experimental \(\sigma_i\) and \(H_i\) are in good agreement with the present model.

It was also found that the \(\sigma_i\) and \(H_i\) of \(\text{MnBi}/\text{Fe-Co}\) composite magnet in Fig. 3(a) and (b) are well fitted to Eqs (13) and (14). Lastly, Eqs (13) and (14) are also validated by the experimental \(\sigma_i\) and \(H_i\) of \(\text{BaM}/\text{Fe-Co}\) composite shown in Fig. 4(a) and (b).

It is noted that a kink in the hysteresis loop becomes more obvious as the \(f^m_h\) decreases, indicating weak or no exchange coupling (not shown in this paper). Therefore, regardless of exchange coupling, the present model can be used to estimate \(\sigma_i\) and \(H_i\) of any powdered hard/soft magnet composite.
Figure 2. The mass fraction ($f_{hm}$) of MnAl dependence of (a) saturation magnetization ($\sigma_s$) and (b) intrinsic coercivity ($H_{ci}$) of MnAl/Fe-Co. The black solid line and red closed circle indicate calculated and experimental data, respectively.

Figure 3. The mass fraction ($f_{hm}$) of MnBi dependence of saturation magnetization ($\sigma_s$) and (b) intrinsic coercivity ($H_{ci}$) of MnBi/Fe-Co. The black solid line and red closed circle indicate calculated and experimental data, respectively.

Figure 4. The mass fraction ($f_{hm}$) of BaM dependence of saturation magnetization ($\sigma_s$) and (b) intrinsic coercivity ($H_{ci}$) of BaM/Fe-Co. The black solid line and red closed circle indicate calculated and experimental data, respectively.
Summary
In summary, we have modified the previously proposed models\(^1,3\) for magnetization (\(M\)) and anisotropy constant (\(K\)) of a hard/soft composite magnet to use mass fraction instead of the volume fraction of hard or soft phase for magnetization (\(\sigma\)) and intrinsic coercivity (\(H_{ci}\)) of powdered hard/soft composite. Our modified equations have been validated by experimental \(\sigma\) and \(H_{ci}\) of Sm\(_2\)Co\(_7\)/Fe-Co, MnAl/Fe-Co, MnBi/Fe-Co, and BaM/Fe-Co composites. Regardless of exchange coupling, the developed equations can be used to predict the \(\sigma\) and \(H_{ci}\) of a powdered hard/soft composite magnet. The present model can provide guidance for the design of exchange coupled hard/soft composite magnets.

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Author Contributions
J.P. derived the equations and fabricated and measured MnAl/Fe-Co, MnBi/Fe-Co, and BaM/Fe-Co samples. C.R., N.P., and J.P.L. provided experimental data points for Sm-Co/Fe-Co. S.G.K. confirmed the equations with theory of magnetism. W.L. and C.J.C. confirmed the equations with their experimental data points. Y.K.H. initiated and directed this research project. J.P. and Y.K.H. wrote the main manuscript text. All authors reviewed the manuscript.

Additional Information
Competing Interests: The authors declare that they have no competing interests.

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