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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 (σ_s) and intrinsic coercivity (H_{ci}) 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 $\text{Sm}_2\text{Co}_7/\text{Fe-Co}$, $\text{MnAl}/\text{Fe-Co}$, $\text{MnBi}/\text{Fe-Co}$, and $\text{BaFe}_{12}\text{O}_{19}/\text{Fe-Co}$ composites for characterization of their σ_s and H_{ci} . 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 $(BH)_{\max} = (B_r)^2/4$ for $H_{ci} > B_r/2$ or $(BH)_{\max} = (B_r - H_{ci})H_{ci}$ for $H_{ci} < B_r/2$ ¹. B_r is the remanent magnetic flux density, and H_{ci} is the intrinsic coercivity, which is mainly controlled by the magnetocrystalline anisotropy constant (K). Therefore, high B_r and H_{ci} are needed for a large $(BH)_{\max}$. In addition, the PM must also have a corresponding high Curie temperature (T_c) 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^{2,3}. Exchange coupling makes full use of high H_{ci} from the hard phase and B_r 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 phase⁴. 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 ($2\delta_w$) of hard magnetic phase for full exchange coupling². Thus, the increasing rate of $(BH)_{\max}$ with the amount of soft phase is limited. Although the previously proposed models^{2,3} predict the magnetization in the unit of emu/cm^3 (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 (σ_s) and H_{ci} 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 σ_s and H_{ci} of $\text{Sm}_2\text{Co}_7/\text{Fe-Co}$, $\text{MnAl}/\text{Fe-Co}$, $\text{MnBi}/\text{Fe-Co}$, and $\text{BaFe}_{12}\text{O}_{19}(\text{BaM})/\text{Fe-Co}$, where Sm_2Co_7 , 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 σ_s and H_{ci} in terms of mass fraction of composite. According to theoretical studies on a two-phase composite magnet, the saturation magnetization² and anisotropy constant³ of a composite can be expressed as:

$$M = f_s M_s + f_h M_h \quad (1)$$

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and

$$K = f_s K_s + f_h K_h, \quad (2)$$

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 σ_s and H_{ci} (per unit mass) of a two-phase magnetic composite using experimentally accessible σ_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/cm³), they can be expressed as:

$$M = \frac{\text{magneticmoment}}{\text{volume}} = \frac{\text{magneticmoment}}{\text{mass}} \cdot \frac{\text{mass}}{\text{volume}} = \sigma \cdot \rho$$

where σ is the saturation magnetization (per unit mass in the unit of emu/g) and ρ is the mass density (in the unit of g/cm³). Therefore, the σ (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:

$$\sigma = \frac{\sigma_h \rho_h f_h + \sigma_s \rho_s f_s}{\rho_h f_h + \rho_s f_s}. \quad (3)$$

H_{ci} due to magnetocrystalline anisotropy⁵ is

$$H_{ci} = \frac{\alpha K}{\sigma \rho}, \quad (4)$$

where α is a constant dependent on the crystal structure and degree of alignment. α is 2 in the case of aligned particles⁶ while for unaligned (random) particles, α can have different values for different crystals (for instance, 0.64 for cubic crystals⁷ 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):

$$H_{ci} = \alpha \frac{(1 - f_h)K_s + f_h K_h}{(1 - f_h)\sigma_s \rho_s + f_h \sigma_h \rho_h}. \quad (5)$$

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

$$H_{ci} = \frac{\sigma_h \rho_h H_h f_h + \sigma_s \rho_s H_s f_s}{\sigma_h \rho_h f_h + \sigma_s \rho_s f_s}, \quad (6)$$

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 σ_s 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.,

$$f_h = \frac{V_h}{V_h + V_s}, \quad (7)$$

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

$$\sigma = \frac{\sigma_h V_h \rho_h + \sigma_s V_s \rho_s}{V_h \rho_h + V_s \rho_s} \quad (8)$$

and

$$H_{ci} = \frac{\sigma_h H_h V_h \rho_h + \sigma_s H_s V_s \rho_s}{\sigma_h V_h \rho_h + \sigma_s V_s \rho_s}, \quad (9)$$

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:

$$\sigma = \frac{\sigma_h \frac{V_h \rho_h}{V_h \rho_h + V_s \rho_s} + \sigma_s \frac{V_s \rho_s}{V_h \rho_h + V_s \rho_s}}{\frac{V_h \rho_h}{V_h \rho_h + V_s \rho_s} + \frac{V_s \rho_s}{V_h \rho_h + V_s \rho_s}} \quad (10)$$

and

$$H_{ci} = \frac{\sigma_h H_h \frac{V_h \rho_h}{V_h \rho_h + V_s \rho_s} + \sigma_s H_s \frac{V_s \rho_s}{V_h \rho_h + V_s \rho_s}}{\sigma_h \frac{V_h \rho_h}{V_h \rho_h + V_s \rho_s} + \sigma_s \frac{V_s \rho_s}{V_h \rho_h + V_s \rho_s}}. \quad (11)$$

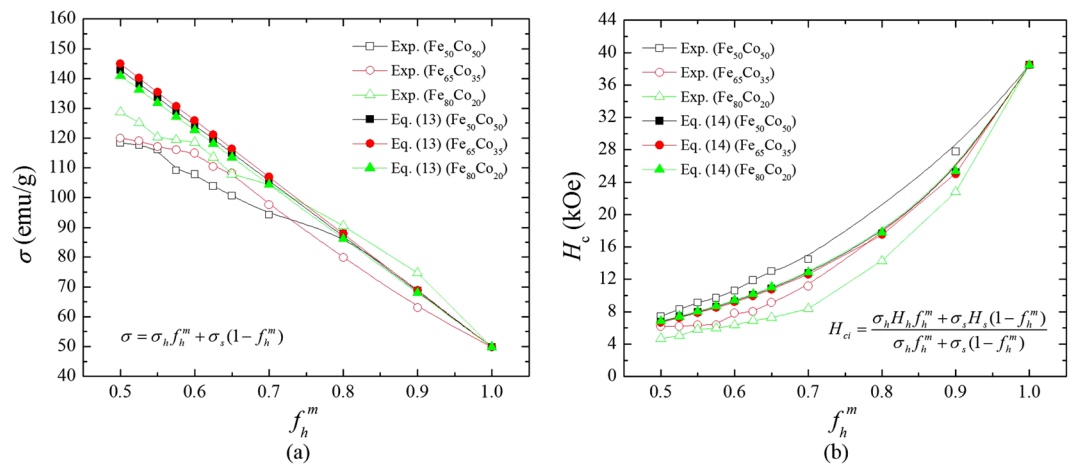


Figure 1. The mass fraction (f_h^m) of Sm_2Co_7 dependence of (a) saturation magnetization (σ_s) and (b) intrinsic coercivity (H_{ci}) of $\text{Sm}_2\text{Co}_7/\text{Fe-Co}$. The open and closed squares indicate experimental and calculated σ_s and H_{ci} , respectively, for $\text{Sm}_2\text{Co}_7/\text{Fe}_{50}\text{Co}_{50}$ composites, circles indicate experimental and calculated σ_s and H_{ci} for $\text{Sm}_2\text{Co}_7/\text{Fe}_{65}\text{Co}_{35}$ composites, and triangles indicate experimental and calculated σ_s and H_{ci} for $\text{Sm}_2\text{Co}_7/\text{Fe}_{80}\text{Co}_{20}$ composites.

The mass fraction of hard (f_h^m) and soft (f_s^m) magnetic phases are

$$f_h^m = \frac{V_h \rho_h}{V_h \rho_h + V_s \rho_s} \text{ and } f_s^m = \frac{V_s \rho_s}{V_h \rho_h + V_s \rho_s}, \quad (12)$$

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

$$\sigma = \sigma_h f_h^m + \sigma_s (1 - f_h^m) \quad (13)$$

and

$$H_{ci} = \frac{\sigma_h H_h f_h^m + \sigma_s H_s (1 - f_h^m)}{\sigma_h f_h^m + \sigma_s (1 - f_h^m)}, \quad (14)$$

respectively.

Eqs (13) and (14) can now be used to estimate the σ_s and H_{ci} of a two-phase magnet by only considering the mass fraction (f_h^m or f_s^m) 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 is 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 σ_s and H_{ci} 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_h^m dependence of σ_s and H_{ci} for $\text{Sm}_2\text{Co}_7/\text{Fe-Co}$ composite with various compositions of Fe-Co. The σ_s decreases linearly as the amount of hard phase (Sm_2Co_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 σ_s from the solid (theoretical) line is getting larger. In Fig. 1(b), experimental H_{ci} 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 σ_s of $\text{MnAl}/\text{Fe-Co}$ composite linearly decreases by increasing the content of hard phase, and the H_{ci} increases by following the developed equation (14). Both experimental σ_s and H_{ci} are in good agreement with the present model.

It was also found that the σ_s and H_{ci} 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 σ_s and H_{ci} 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_h^m 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 σ_s and H_{ci} of any powdered hard/soft magnet composite.

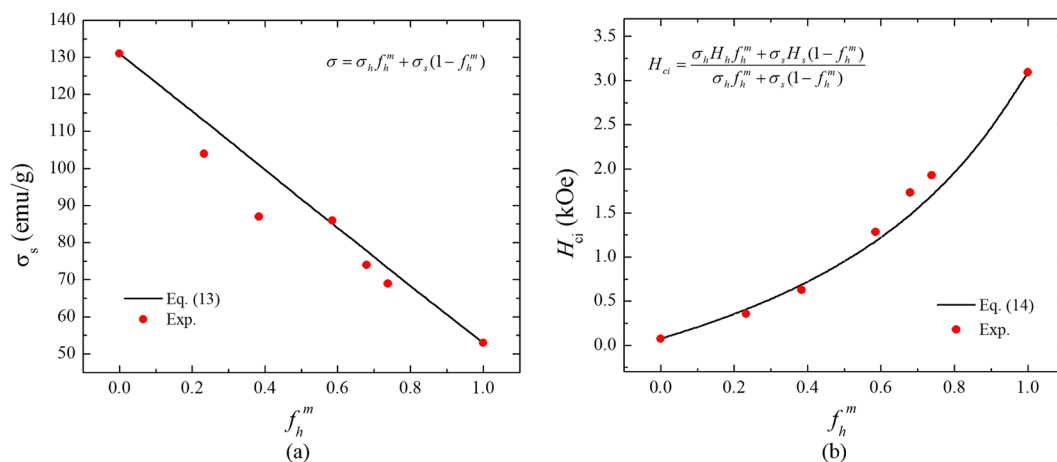


Figure 2. The mass fraction (f_h^m) of MnAl dependence of (a) saturation magnetization (σ_s) and (b) intrinsic coercivity (H_{ci}) of MnAl/Fe-Co. The black solid line and red closed circle indicate calculated and experimental data, respectively⁸.

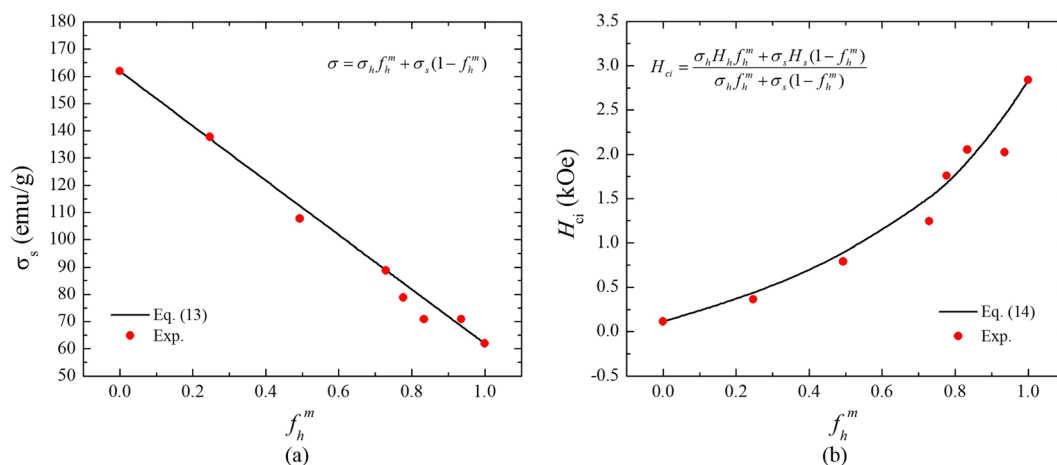


Figure 3. The mass fraction (f_h^m) of MnBi dependence of saturation magnetization (σ_s) and (b) intrinsic coercivity (H_{ci}) of MnBi/Fe-Co. The black solid line and red closed circle indicate calculated and experimental data, respectively.

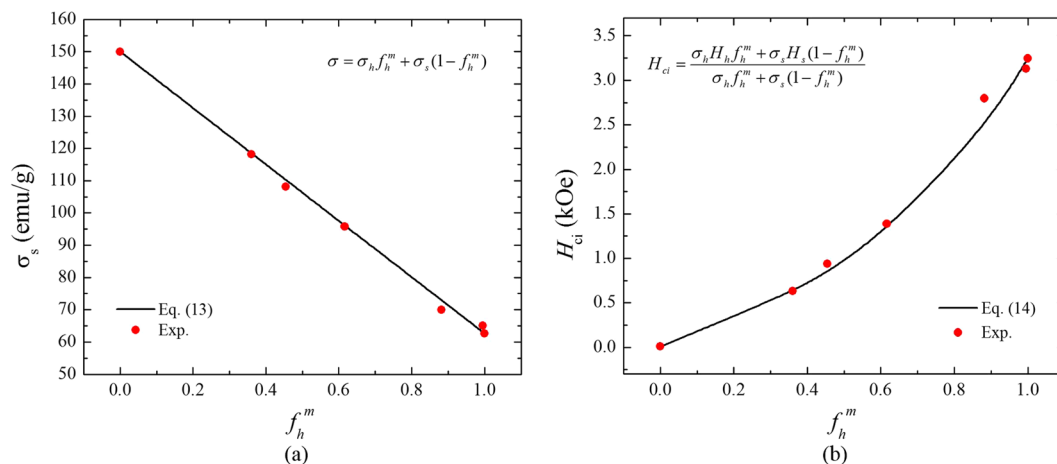


Figure 4. The mass fraction (f_h^m) of BaM dependence of saturation magnetization (σ_s) and (b) intrinsic coercivity (H_{ci}) of BaM/Fe-Co. The black solid line and red closed circle indicate calculated and experimental data, respectively.

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 (σ_s) and intrinsic coercivity (H_{ci}) of powdered hard/soft composite. Our modified equations have been validated by experimental σ_s and H_{ci} of $\text{Sm}_2\text{Co}_7/\text{Fe-Co}$, $\text{MnAl}/\text{Fe-Co}$, $\text{MnBi}/\text{Fe-Co}$, and $\text{BaM}/\text{Fe-Co}$ composites. Regardless of exchange coupling, the developed equations can be used to predict the σ_s 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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