

# First-Order-Reversal-Curve Analysis of Permanent Magnet Materials

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Rare-earth permanent magnet materials are indispensable elements in many electronic devices such as electrical motors, hybrid vehicles, portable communications devices, etc. The magnets have major influence on the size, efficiency, stability, and cost of these devices and systems. The development of rare-earth magnets in the 1970s profoundly influenced the application of permanent magnetic materials owing to their large energy product (i.e.,  $BH_{max}$ ) and increased volume efficiency. The rare-earth magnets that have been most extensively studied and developed are SmCo and NdFeB<sup>1</sup>.

Over the last couple of decades there's been interest in the development of nanostructured magnets, and exchange-coupled nanocomposite alloys with co-existing soft and hard phases because of the coercivity enhancement that is obtained at the single-domain size (nanometer scale)<sup>2,3</sup>. The magnetic characterization of such materials is usually made by measuring a hysteresis loop, however it is not possible to obtain information of interactions or coercivity distributions from the hysteresis loop alone. First-order-reversal-curves (FORCs) provide a means for determining the distribution of interaction fields between magnetic particles.

Vibrating sample magnetometers (VSM) are the most commonly employed technique for measuring the magnetic properties of permanent magnet materials because measurements can be performed on solids, powders, single crystals, thin films, nanostructures and liquids, and because measurements can be performed over a broad range of temperatures (4 K to 1273 K) and magnetic fields employing either electromagnets (3 T)<sup>4,5</sup> or high-field superconducting magnets (16 T)<sup>6,7</sup>. In this paper we will discuss the FORC measurement and analysis technique and present results for three permanent magnet samples: SmCo nanoparticles, AlNiCo, and a sample consisting of a mixture of soft (ferrite) and hard (SmCo) phases.

## The VSM Technique

If a material is placed within a uniform magnetic field  $H$ , a magnetic moment  $m$  will be induced in the sample. In a VSM, a sample is placed within suitably placed sensing coils, and is made to undergo sinusoidal motion, i.e., mechanically vibrated. The resulting magnetic flux changes induce a voltage in the sensing coils that is proportional to the magnetic moment of the sample<sup>8</sup>. The magnetic field may be generated by an electromagnet or a superconducting magnet. Variable temperatures from cryogenic to high temperatures may be achieved using either cryostats or furnace assemblies.

A VSM measures magnetic moment  $m$ , however in some cases the quantity of interest is the materials magnetization  $M$ . The magnetization  $M$  (in cgs units) can be expressed in terms of the mass (emu/g) or volume magnetization (emu/cc) and is the moment  $m$  divided by the sample mass or volume, respectively. A VSM is most commonly used to measure a materials major hysteresis or  $M(H)$  loop. In some cases it is preferred to present the magnetization data in terms of the magnetic induction  $B$  which in cgs units has the unit gauss (G). The relation between  $M$  and  $B$  is:  $B(G) = H + 4\pi M$ , where  $M$  is the volume magnetization (emu/cc).

## VSM Measurements of Permanent Magnet Materials

Figure 1 shows the initial magnetization curve, and major  $M(H)$  loop<sup>4</sup> for a ~1 mg SmCo nanoparticles powder sample. The most common parameters extracted from the hysteresis loop that are used to characterize the magnetic properties of permanent magnet materials include: the saturation magnetization  $M_s$  (the magnetization at maximum applied field), the remanence  $M_r$  (the magnetization at zero applied field after applying a saturating field), the coercivity  $H_c$  (the field required to demagnetize the sample), and the energy product  $BH_{max}$ , which is the magnetic field strength at the point of maximum energy product of a magnetic material.  $BH_{max}$  is obtained from the 2<sup>nd</sup> quadrant or demagnetization curve of the material (i.e., upper left quadrant in figure 1), where the curve is recorded starting at remanence  $M_r$  and ending at the coercivity  $H_c$ .

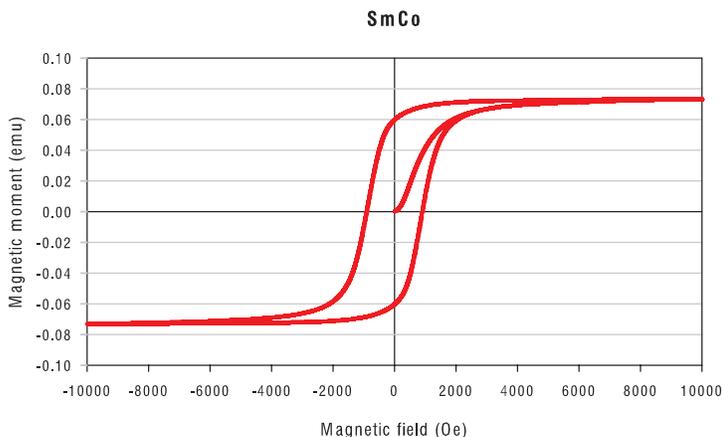


Figure 1: Initial magnetization and major hysteresis loop for a SmCo nanoparticles powder sample. The data are presented in terms of magnetic moment (emu) versus applied magnetic field (Oe).

## First-Order-Reversal-Curves (FORCs)

More complex magnetization curves covering states with field and magnetization values located inside the major hysteresis loop, such as first-order-reversal-curves (FORCs) can give additional information that can be used for characterization of magnetic interactions. A FORC is measured by saturating a sample in a field  $H_{sat}$ , decreasing the field to a reversal field  $H_a$ , then sweeping the field back to  $H_{sat}$  in a series of regular field steps  $H_b$ . This process is repeated for many values of  $H_a$  yielding a series of FORCs. The measured magnetization at each step as a function of  $H_a$  and  $H_b$  gives  $M(H_a, H_b)$ , which is then plotted as a function of  $H_a$  and  $H_b$  in field space. The FORC distribution  $\rho(H_a, H_b)$  is the mixed second derivative, i.e.,  $\rho(H_a, H_b) = -\partial^2 M(H_a, H_b) / \partial H_a \partial H_b$ , and a FORC diagram is a contour plot of  $\rho(H_a, H_b)$  with the axis rotated by changing coordinates from  $(H_a, H_b)$  to  $H_c = (H_b - H_a)/2$  and  $H_u = (H_b + H_a)/2$ , where

$H_u$  represents the distribution of interaction fields, and  $H_c$  represents the distribution of switching fields.

Figure 2 shows a series of FORCs<sup>4</sup> for a SmCo nanoparticles powder sample, and figure 3 shows the corresponding FORC diagram<sup>9</sup>. From the major hysteresis loop shown in figure 1, the coercivity  $H_c = 900$  Oe. From the FORC diagram shown in figure 3 there is a single maximum in the distribution that is centered around  $H_c$  (900 Oe), the distribution of switching fields extends over several hundreds of Oe because different particles switch at different applied field strengths. The small spread of the distribution in the  $H_u$  direction is an indication that the interaction between particles is relatively weak.

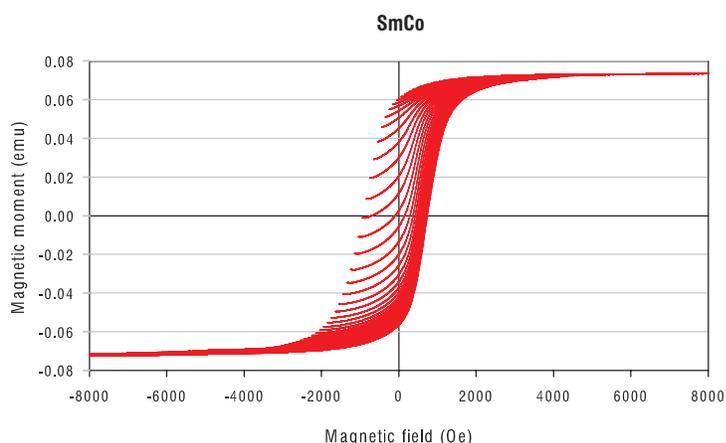


Figure 2: FORCs for a SmCo nanoparticles powder sample.

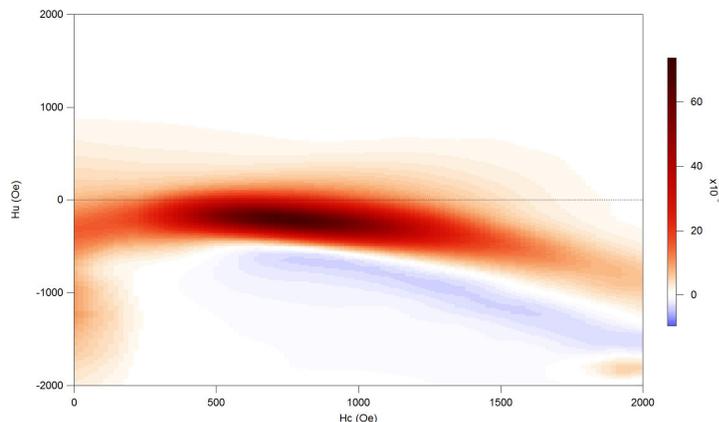


Figure 3: FORC diagram for a SmCo nanoparticles powder sample.

Figures 4, 5, and 6 show the initial magnetization and major hysteresis loop, a series of FORCs<sup>4</sup>, and the corresponding FORC diagram<sup>9</sup>, respectively, for an AlNiCo permanent magnet. From the major hysteresis loop shown in figure 4, the coercivity  $H_c = 600$  Oe, and from the FORC diagram shown in figure 6 there is a single maximum in the distribution that is centered around  $H_c$  (600 Oe). As for the SmCo sample note, however, the distribution of switching fields extends over a few hundred Oe owing to particles switching at different applied field strengths. In this case the spread in the  $H_u$  is larger and indicates a larger interaction between the particles. It is clear from these examples that the FORC measurement protocol and analysis provide additional information regarding irreversible magnetic interactions or processes in permanent magnet materials that cannot be obtained from the standard hysteresis loop measurement alone.

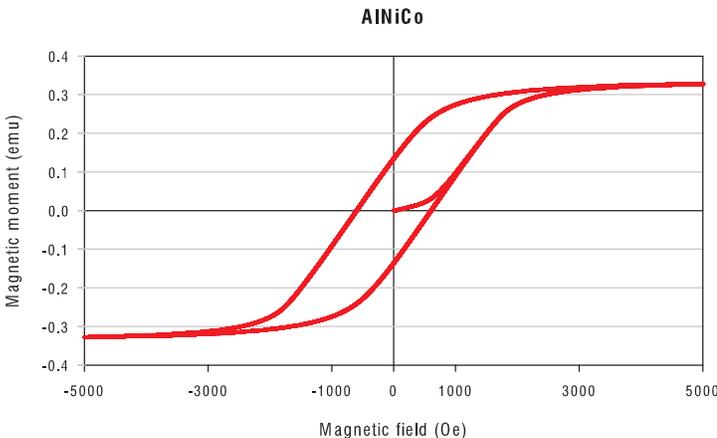


Figure 4: Initial magnetization and major hysteresis loop for AlNiCo. The data are presented in terms of magnetic moment (emu) versus applied magnetic field (Oe).

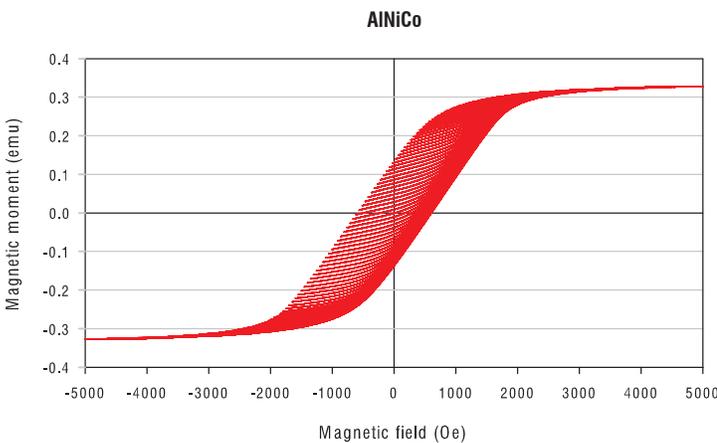


Figure 5: FORCs for AlNiCo sample.

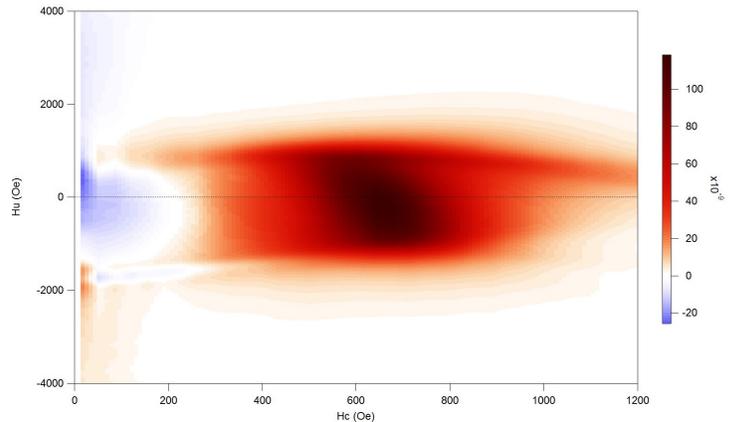


Figure 6: FORC diagram for AlNiCo.

For exchange coupled alloys that contain both magnetically soft and hard phases, FORC analysis provides for differentiation of the phases<sup>2,3</sup>. To demonstrate this, FORC analysis was performed on a mixture of soft ferrite and hard SmCo powders. From the hysteresis loop measurements of the samples separately, the coercivities were 275 Oe (ferrite) and 900 Oe (SmCo). The hysteresis loop for the mixed sample is shown in figure 7, and is clearly dominated by the hard (SmCo) phase ( $H_c = 900$  Oe). From the hysteresis loop alone it is not possible to differentiate between the soft and hard phases.

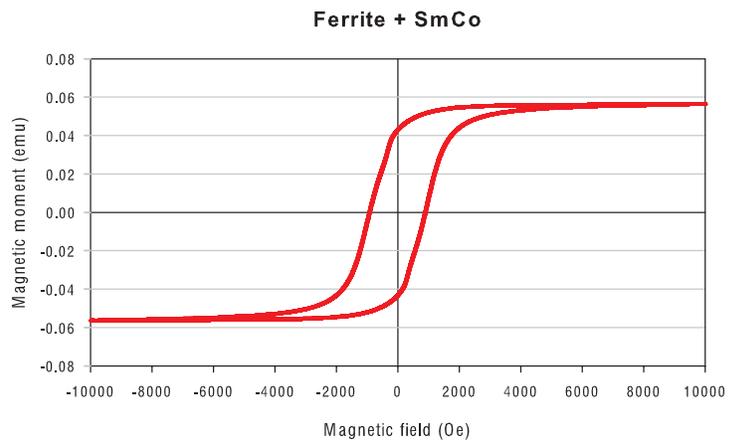


Figure 7: Major hysteresis loop for a mixture of ferrite and SmCo powders.

Figures 8, 9, and 10 show a series of FORCs<sup>4</sup> and the FORC diagram<sup>9</sup> for the mixed sample. For clarity, contours have been added to the second FORC diagram. The soft (ferrite) and hard (SmCo) phases are clearly differentiated, demonstrating the utility of FORC analysis for characterizing the magnetic properties of magnets containing both soft and hard phases.

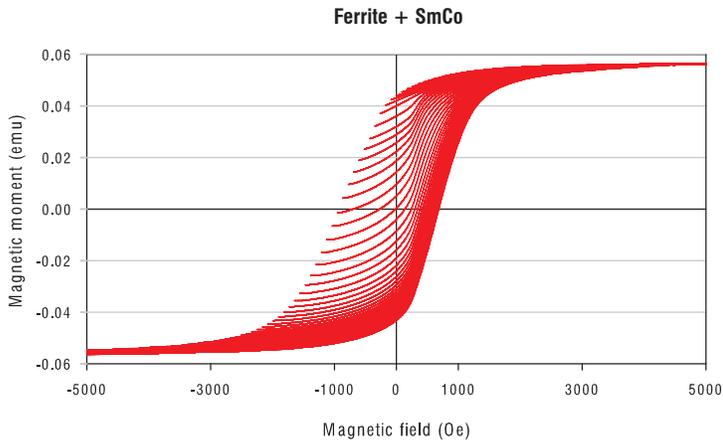


Figure 8: FORCs for a mixed ferrite (soft) and SmCo (hard) sample.

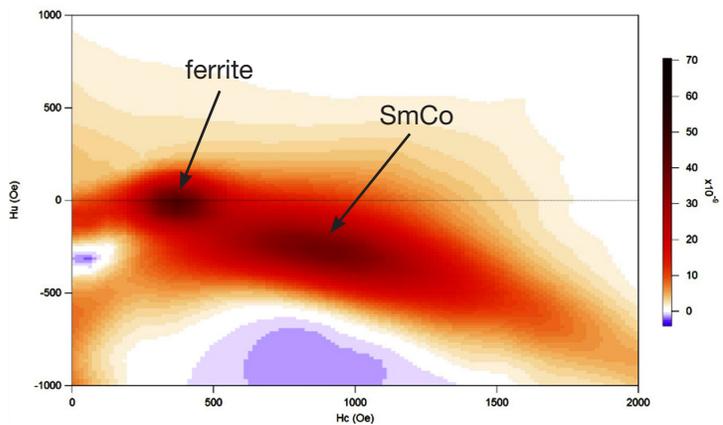


Figure 9: FORC diagram for a mixed ferrite (soft) and SmCo (hard) sample.

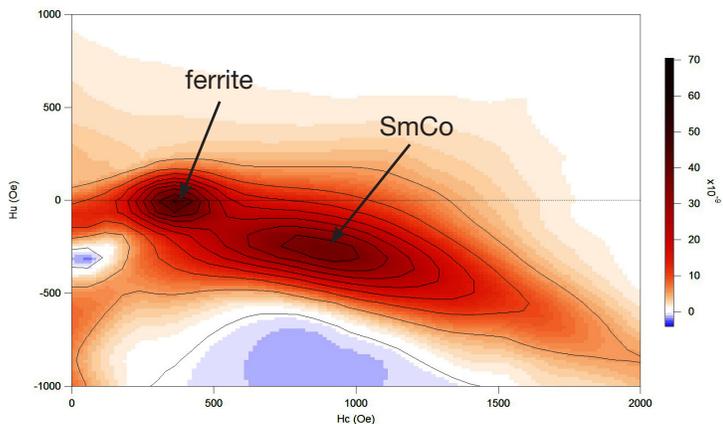


Figure 10: FORC diagram with contours for a mixed ferrite (soft) and SmCo (hard) sample.

## Conclusion

VSMs are ideally suited to investigate the magnetic properties of permanent magnet materials. While measurements of a materials major hysteresis loop yield several desirable quantities of interest: coercivity ( $H_c$ ), saturation magnetization ( $M_s$ ), remanence ( $M_r$ ), and energy product  $BH_{max}$ , they cannot be used to extract information regarding interactions and coercivity distributions that reveal insight into the relative proportions of reversible and irreversible components of the magnetization in a material. To accomplish this one must use the FORC measurement technique and subsequent analysis to produce the FORC diagram, which reveals data not readily discerned from the hysteresis loop.

## References

- 1 B. C. Dodrill, "Characterizing permanent magnet materials with a vibrating sample magnetometer", Magnetics Business & Technology, 12, Fall 2012
- 2 M. Pan, P. Zhang, H. Ge, N. Yu, Q. Wu, "First-order-reversal-curve analysis of exchange-coupled SmCo/NdFeB nanocomposite alloys", Journal of Magnetism and Magnetic Materials, 361, 219, 2014
- 3 P. Chen, C. Yang, S. Chang, M. Lee, N. Tang, S. Yen, Y. Tseng, "Soft and hard natures of  $Nd_2Fe_{14}B$  permanent magnet explored by first-order-reversal-curves", Journal of Magnetism and Magnetic Materials, in press, 2014
- 4 Lake Shore Cryotronics, USA
- 5 Microsense, USA
- 6 Quantum Design, USA
- 7 Cryogenic Limited, UK
- 8 S. Foner, "Versatile and sensitive vibrating sample magnetometer", Review of Scientific Instruments, 30, 548, 1959
- 9 R. J. Harrison and J. M. Feinberg, "FORCinel: An improved algorithm for calculating first-order reversal curve distributions using locally weighted regression smoothing", Geochemistry, Geophysics, Geosystems, 9, 11, 2008