

Composition Dependence of the Magnetic Properties of Bonded Magnets of Strontium Hexaferrite-Polyvinyl Chloride

J. Matutes-Aquino, D. Ríos-Jara, O. Ayala-Valenzuela, P. Sifuentes-Gallardo, L. F. Ramos de Valle, O.S. Rodríguez Fernández.

Abstract

A systematic study of the magnetic properties of strontium ferrite-polyvinyl chloride plastoferrites as a function of composition is presented. Plastoferrites were manufactured by hot compression of a mixture of commercial powders of strontium ferrite ($\text{SrFe}_{12}\text{O}_{19}$) and polyvinyl chloride. In order to explain the dependence of the volumetric density and the saturation magnetization with composition, an additive model for these properties is considered. The intrinsic coercivity shows a decrease with increasing strontium ferrite content, which is interpreted as a magnetizing-like effect due to the increasing interaction between the magnetic particles. The maximum energy products are lower than 1 MGOe and increase with the second power of the strontium ferrite content. The best maximum energy product and specific saturation magnetization were obtained for the plastoferrite with a 85 weight percentage of ferrite.

Introduction

The problem of embedding magnetizable particles during or after the processing of a polymer matrix is very interesting because of the actual and future applications of these composite materials (1).

Many sintered magnets are brittle, hard and difficult to machine to their final forms. Plastoferrites are manufactured by consolidating magnetic powder with a polymeric matrix, which makes machining easy and sometimes even unnecessary.

According to the application, the plastic matrix can be a thermosetting, as the epoxy resin used in compression molded magnets, a thermoplastic, like nylon used in injection molded magnets or an elastomer, used for extruded magnets. Elastomeric materials represent an important class of engineering materials that are widely used to make components of structures, machinery, and devices for vibration and noise control.

Elastomeric materials with embedded metallic particles are used in applications such as flexible, electrically and magnetically conductive interfaces and tapes (2), flexible permanent magnets (3). There has been recent interest in creating controllable materials based on elastomers that are loaded with magnetically conductive particles (4, 5). Such controllable elastomers promise to have more functionality than conventional elastomers and therefore could be a link that brings the applications of modern control technologies, intelligent structures, and smart materials to a very broad industrial area. From the scientific point of view exist only a few references. Jolly and coworkers (6) report on the magnetoviscoelastic behavior of composites of elastomers and iron particles. The main disadvantage in all of them is the reduction in magnetic properties compared with a magnet made with 100% magnetic material (7-9). Plasticized PVC offers a good option to incorporate high levels of particles, which are needed to obtain good magnetical properties maintaining a good balance on mechanical properties.

Plastofemtes are used in many applications, such as holding magnets for refrigerator catches, correction magnets for television tubes, fractional-horsepower motors and dynamos, separators (10- 12).

Table 1. PVC Formulation in Parts Per Hundred Parts of Resin (phr).

Ingredients	phr
PVC	2000
Lead compound (thermal stabilizer)	5.5
Plasticizer	50
Zinc stearate (lubricant)	4.0

In this work the magnetic characterization at room temperature of isotropic bonded magnets of strontium ferrite ($\text{SrFe}_{12}\text{O}_{19}$)-polyvinyl chloride (PVC) as a function of composition is presented.

Experimental procedure

PVC compound Preparation: Suspension poly (vinyl chloride) (PVC Corvic S71/102), with a K value of 71 was supplied by ICI Ltd., U.K.; di 2-ethyl hexyl phthalate (DOP) was used as plasticizer. The PVC formulation used is shown in Table 1; values are in parts per hundred parts of resin (phr). The compounds were dry blended in a Henschel intensive mixer. The resin was preheated to 70°C, after which the lubricant and the thermal stabilizer were added: finally, the plasticizer was added at 90°C. A total mixing time of 20 minutes was chosen in order to achieve good mixing.

PVC- Strontium Ferrite Composite Preparation: WC dry blends were prepared at 180°C at 60 rpm for 5 minutes using a Brabender mixing chamber equipped with roller mixers. Once the WC compound was fused the magnetic particles were added, the melt temperature was 185°C. Different plastic bonded compounds were prepared, in which the magnetic particle concentration varied from 10 to 85 wt%. The obtained compounds were chopped and then compression molded for 5 minutes at 180°C (platen temperature) using a pressure of 10 MPa. Dumbbell specimens were cut using a BS003 A-2 die cutter: 2 mm-thick samples were obtained.

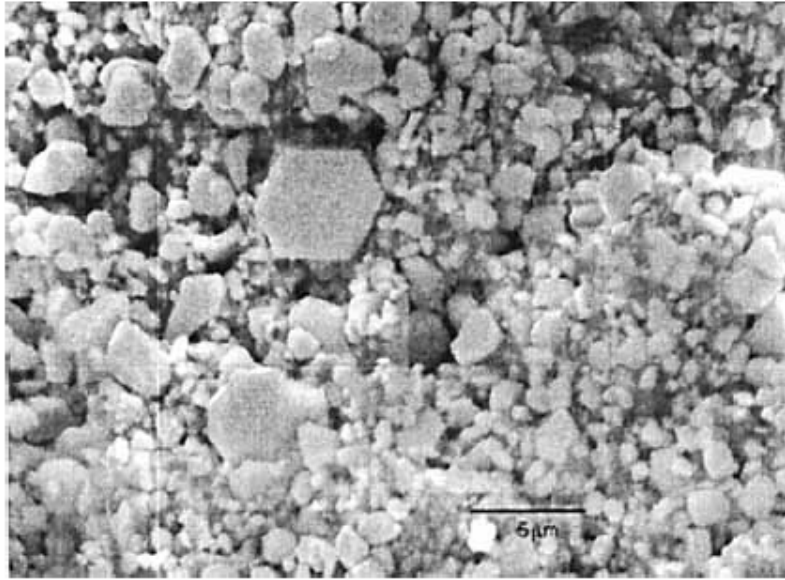


Fig. 1. Scanning electron micrograph of the microstructure of the plastoferrite (0.85 mass fraction of strontium ferrite) with a distribution of strontium ferrite hexagonal plates in polyvinyl chloride.

Magnetic Measurements: The room temperature magnetic characterization was made with a vibrating sample magnetometer model 9600 from LDJ ELECTRONICS with a maximum applied magnetic field of 16 kOe. Samples 3 X 3 x 3 mm were used for the magnetic measurements of the plastoferrites with the magnetic field applied along one cube axis. A cylinder 8 mm long and 4 mm diameter was used for the magnetic measurements of the strontium ferrite powder with the magnetic field applied perpendicular to the cylinder axis. No further corrections for the demagnetizing field effect were made.

Results and discussion

Figure 1 shows the microstructure of the plastoferrite with a distribution of strontium ferrite hexagonal plates in polyvinyl chloride.

Assuming an additive model, the volume density and the saturation magnetization result from the weighted addition of the contributions of each component.

In this additive model the contribution to the volume density of each component is independent, therefore the volume density of the composite varies linearly with the volume fraction of strontium ferrite according to the formula:

$$\rho = \rho_p + (\rho_f - \rho_p)f_{vf} \quad (1)$$

where ρ is the volumetric density of the plastoferrite, ρ_p is the volumetric density of the plastic matrix, ρ_f is the volumetric density of pure dense strontium ferrite and f_{vf} is the volume fraction of strontium ferrite in the plastoferrite.

Figure 2 shows the measured volume densities of the plastoferrites as a function of the volume fraction of strontium ferrite. These densities values are described by the straight line corresponding to Eq 1.

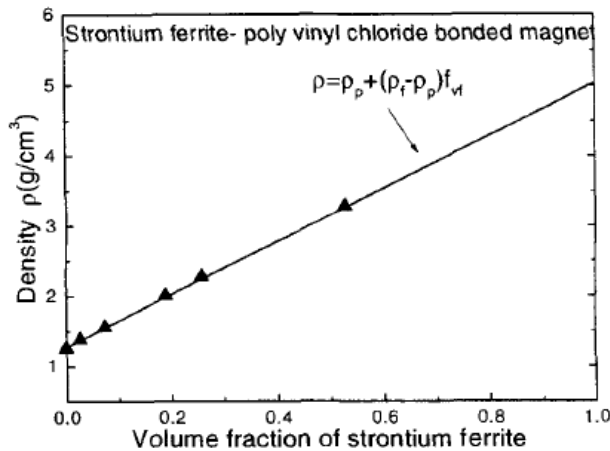


Fig. 2. Volume density as a function of volume fraction of strontium ferrite in the plastoferrites.

In this Equation the plastic matrix density, $\rho_p = 1.256 \text{ g/cm}^3$, was experimentally determined and the strontium ferrite density, $\rho_f = 5.107 \text{ g/cm}^3$, was calculated using the total atomic mass of the hexagonal unit cell and the lattice parameters $a = 0.58836 \text{ nm}$ and $c = 0.23037 \text{ nm}$ as determined by neutron diffraction (13).

Figure 3 shows the initial magnetization curves and the hysteresis loop curves for the different plastoferrite compositions. It can be observed that the magnetization increases with the increasing content of strontium ferrite. From these curves the specific saturation magnetization, in emu/g, and the intrinsic coercivity, in kOe, were determined.

Only the strontium ferrite component contributes to the magnetization, and it can be shown that the specific saturation magnetization, in emu/g, presents a nonlinear dependence with the volume fraction of strontium ferrite according to Eq 2:

$$\sigma_s = \frac{\rho_f \sigma_{sf} f_{vf}}{[(\rho_f - \rho_p)f_{vf} + \rho_p]} \quad (2)$$

where σ_s and σ_{sf} are the specific saturation magnetization of the plastoferrite and the pure strontium ferrite respectively, and f_{vf} is the volume fraction of strontium ferrite. However, it is more simple to consider the dependence with the mass fraction because in this case it is expected a linear dependence of the specific saturation magnetization, in emu/g, with the mass fraction of strontium ferrite according to Eq 3:

$$\sigma_s = \sigma_{sf} f_{mf} \quad (3)$$

Figure 4 shows the variation of the specific saturation magnetization of the plastoferrites with the mass fraction of strontium ferrite. It can be observed that the experimentally determined values of saturation magnetization for different plastoferrite

compositions fit to the expected linear dependence with the mass fraction of strontium ferrite.

Figure 5 shows the intrinsic coercivity of plastroferrites as a function of the mass fraction of strontium ferrite.

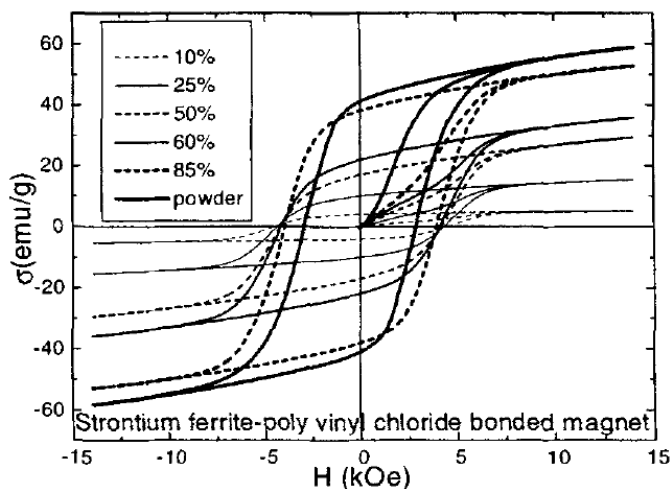


Fig. 3. Hysteresis loops of plastroferrites and strontium ferrite powder.

A straight line could be fitted to the experimental values to describe the reduction of intrinsic coercivity as the mass fraction of strontium ferrite increases. The mean distance between the strontium ferrite plates decreases with the increasing of the strontium ferrite mass fraction. For that reason this system becomes very interesting for studying the interactions between magnetic particles. The interaction between magnetic particles is a complex many body problem. Different authors have used the Wohlfarth relation and the Henkel plots to study the presence of interaction in concentrated magnetic systems. They have found magnetizing and demagnetizing effects due to the interaction between neighboring particles or grains (14). In our case, the behavior of the intrinsic coercivity is qualitatively understood in terms of the magnetic interactions

between the strontium ferrite particles as they approach to each other when increasing the mass fraction of strontium ferrite. It seems that on the average, the magnetic field from one particle acting on another particle helps the applied magnetic field reduce the intrinsic coercivity of the material. It is a magnetizing-like effect.

The σ -H curves can be converted into B-H curves with the help of the fundamental relationship between the magnetic vectors, $B = H + 4\pi M$, where M is the volume magnetization in emu/cm³. The specific magnetization σ , in emu/g, and the volume magnetization M, in emu/cm³, are related with the volume density ρ of the material, in g/cm³, through the equation $M = \sigma\rho$. The second quadrant part of a B-H hysteresis loop is called the demagnetization curve. For a specific application the operation point of a permanent magnet will be somewhere in the demagnetization curve. The lefthand-side curves shown in Fig. 6 correspond to demagnetization curves of the studied plastoferrites. Contrary to the intrinsic coercivity shown in Fig. 5, in this case the coercive force increases with the content of strontium because to zero the flux density B at the coercive force, it is necessary to compensate the magnetization M with the applied field H, and therefore the plastofenite with a higher saturation magnetization (with a higher ferrite content) would require a higher magnetic field H to reach this condition.

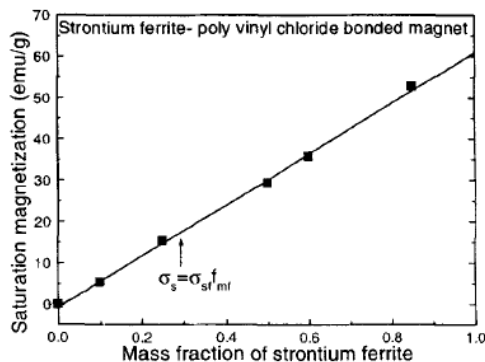


Fig. 4. Specific saturation magnetization as a function of mass fraction of strontium ferrite.

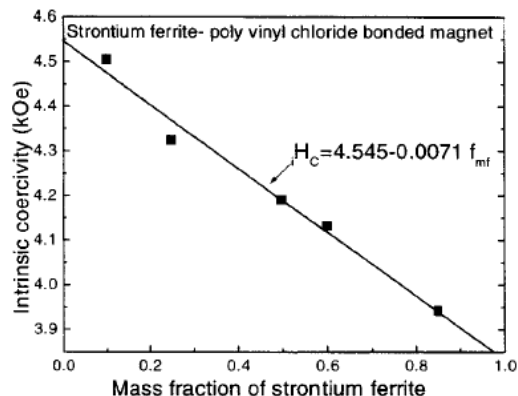


Fig. 5. Intrinsic coercivity as a function of mass fraction of strontium ferrite.

The energy product (BH) is obtained by multiplying the flux density (B) by the magnetic field intensity (H) for each point of the demagnetization curve. This important quantity is proportional to the stored energy in a permanent magnet. The right-hand curves in Fig. 6 correspond to the energy product of the studied plastoferrites as a function of the flux density B. Every (BH) energy product curve has a maximum value $(BH)_{max}$ for a given value of the flux density. Often it is desirable that a permanent magnet work at this maximum value, if the maximum stored energy density is to be used. However, other considerations like a higher coercive force or a higher saturation flux density can define a different operating point (15, 16).

Figure 7 shows a quadratic dependence of the $(BH)_{max}$ values with the volume fraction of strontium ferrite. As expected, a second order equation could be perfectly fitted to the experimental data so that the maximum energy product increases with the second power of the volume fraction of strontium ferrite (7).

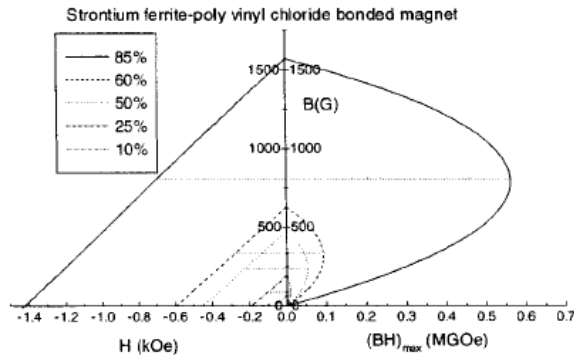


Fig. 6. Demagnetization and $B \times H$ energy product curves for different compositions.

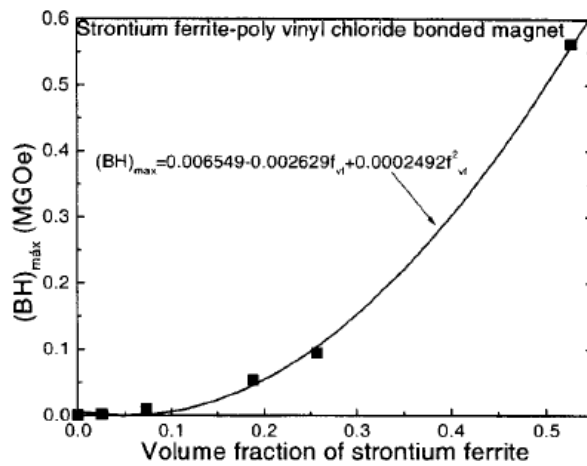


Fig. 7. Variation of the maximum energy product as a function of the volume fraction of strontium ferrite.

From these results it becomes clear that in order to optimize the magnetic properties (higher magnetization and higher $(BH)_m$) the objective should be to maximize the volume fraction of magnetic powder used. However for compression it is not recommended to use more than 85 wt% percentage of ferrite molding while still maintaining adequate flow characteristics for molding the compound and sufficient particle bonding for good mechanical properties (7).

Conclusions

1. The experimental composition dependence of the volumetric density and the specific saturation magnetization of plastoferrites could be described with a linear additive model of these properties.

2. The intrinsic coercivity decreases linearly with increasing strontium ferrite concentration, which is interpreted as a magnetizing-like effect due to the increasing magnetic interaction between the strontium ferrite particles as the mean interparticle distance decreases.

3. As expected for isotropic plastoferrites, maximum energy products lower than 1 MGOe were obtained and a quadratic dependence of the maximum energy product values with the volume fraction of strontium ferrite was found.

References

1. H. Stablein, "Hard Ferrites and Plastoferrites," in E. P. Wohlfahrt, ed., *Ferromagnetic Materials*, Vol. 3, 441 (1982).
2. Zoppi et al., *J. Polymer Sci: Part A: Polymer Chemistry*, 52, 1001 (1994).
3. M. Hanasaka, *Roc. Inter-noise 94*, p. 693.
4. B. C. M~xioazn d C. R. Jolly, 4th Annual Workshop: Advances in Smart Materials for Aerospace Applications, NASA Langley Research Center (1995).
5. H. Banno and K. Ogura, *J. Ceramic Soc. Japan*, 100 (41), 551 (1992).
6. M. R Jolly, J. D. Carlson, B. E. Mmioz, and T. A. Bullions, *J. Intelligent M a t Sus. and Structures*, 7,613 (1996).
7. P. Campbell, *Permanent Magnet Materials and Their Applications*, p. 51, Cambridge University Press (1994).

8. B. T. Shirk, "High Energy Bonded Ferrite Powders-A New Look at an Old Friend," in Polymer Bonded Magnets 96, Atlanta [April 1996].
9. R. A. McCurrie, Ferromagnetic Materials: Structure and Properties, p. 245, Academic Press (1994).
10. P. D. Hinderaker, Machine Design, 94 (1976).
11. G. Samow, Ekketroanzeigez-, 26, No. 22,453 (1973).
12. J. J. Badner, J. Applied Physics, 49, 1788 (1978).
13. K. Kimura, M. Ohgaki, K. Tanaka, H. Morikawa, and F. Marumo, J. Solid State Chemistry, 87, 186 (1990).
14. F. P. Missell, M. Virginia, P Altoe. and D. Comejo. Proc. III Latin American Workshop on Magnetism, Magnetic Materials and their Applications, p. 228, World Scientific Co. Re. Ltd. (1996).
15. B. D. Cullity, Introduction to Magnetic Materials, p. 560, Addison-Wesley Publishing Co. (1972).
16. R. J. Parker, Advances in Pennanent Magnetism p. 37, John Wiley & Sons (1990).