Rare-Earth Fe–B Powder Coating for Improvements in Corrosion Resistance, Flux Aging Loss, and Mechanical Strength of Bonded Magnets

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Coating formulations have been investigated for rare-earth Fe–B (RE Fe–B) type melt spun magnetic powders, to provide them with enhanced corrosion and oxidation resistance when exposed to aggressive environments. The coating formulations consist of an epoxy binder-based system and specialty additives. Results with two RE Fe–B powder types using these new formulations show between 19 °C and 26 °C enhancements in their operating temperatures for comparable flux aging loss, but now with better magnetic property retention at high temperatures and better adhesion between powder and organic coating. Optimal flux aging loss was found to result after curing the compacts within 180 °C to 200 °C for 30 to 60 min at a compaction pressure of 690 MPa (7 tons cm^{-2}), although higher pressures resulted in more severe magnetic losses but higher bending strengths. The new formulations yield magnets whose magnetic properties are less affected by curing at these typical high temperatures.

Index Terms—**Organic additives, oxidation resistance, powder coatings, rare-earth Fe–B magnets.**

I. INTRODUCTION

POLYMER bonded isotropic rare-earth permanent magnets are increasingly used in a wide variety of electric motors and electronic devices. The criteria for selecting the exact type of magnetic material are strongly influenced by the operation conditions demanded by a given application. The polymer binder used for making polymer bonded magnets must be able to provide sufficient mechanical strength to hold magnet powder together and maintain the designed shape to specification up to the intended operation temperature, and to sustain that operation temperature without softening, deforming or breaking. The magnetic material must provide sufficient flux to sustain the desired properties at the operation temperature without substantial or irreversible loss of magnetization. Its stability to heat and its resistance to corrosive and oxidative environments affect the magnetic material's ability to retain magnetic flux over certain temperatures and time durations; this flux aging loss ultimately determines the magnet's utility. The measure of successful enhancement to such bonded magnets is that oxidative or corrosive degradation of the constituent materials and the consequent change in overall magnetic properties is minimized. A solvent or liquid coating process is an effective manner in which each organic component can be blended with the magnetic particles. The application of a low-viscosity, volatile carrier fluid containing dissolved low-molecular weight binder components and various additives ensures the fast and simple production of compression moldable, liquid-coated, rare-earth Fe–B (RE Fe–B) powders for high-temperature bonded magnet applications.

New materials are being added to polymeric systems called "nanofillers" or "nanoadditives" and are showing remarkable improvements in a system's thermal stability, mechanical strengths, oxygen permeability, and processability. These additives are primarily inorganic and consist of a silica-based structure with organic functionalities.

By incorporating coupling agents and other specialty additives with the magnetic powder and the epoxy components, additional oxidation and corrosion prevention, enhanced adhesion and dispersion between the filler and matrix components have been achieved [1]–[3]. The purpose of this paper is to report the benefits of utilizing these particular additives on magnet properties.

II. EXPERIMENTAL PROCEDURES

Two uncoated magnetic powders used in this study were of the Nd–Fe–B 2:14:1 single-phase type, and had the general formulations: *Powder A*: $\mathrm{Nd_{12}Fe_{77}Co_5B_6}$ with $\mathrm{B_r} = 900$ mT, $\mathrm{H_{ci}} = 780\ \mathrm{kA\,m^{-1}}$; *Powder B*: $\mathrm{Nd_{12}Fe_{80}Nb_2B_6}$ with $\mathrm{B_r} = 846$ mT, $\mathrm{H_{ci}} = 947\ \mathrm{kA\,m^{-1}}$. The difference in microstructure between these two powders results in Powder B having a more brittle flake than Powder A. This is manifested in Fig. 1, where the coated Powder B exhibits a stronger dependence on compaction pressure. The steep linear rise in density at low pressure signifies a material with low yield strength [4]. This fact allows Powder B to achieve slightly higher densities than Powder A, but it will be more susceptible to oxidation due to the creation of fresh surfaces on the coated powders.

These powders were coated with an epoxy-based binder system that comprises either a novolac epoxy or a bisphenol A-type epoxy resin, a dicyandiamide hardener, a cure accelerator, a zinc stearate lubricant, and specialty commercial additives. Of these additives a neoalkoxy-based organotitanate coupling agent with pyrophosphato functionality aids in the cross-linking of the cure reaction and improves compatibility of the binder components with the RE Fe–B powders, which provides efficient protection of the powders in the magnet. Kenny *et al.* [5] discussed the benefits of a montmorillonite

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organoclay present in a bisphenol A epoxy resin at a concentration of 10 wt%. They observed that the organoclay improved the thermal stability and acted as a catalyst for the cure reaction. It is because of this and similar research that we chose to employ the organoclay in our coating system. Our research has shown that an alkyl quaternary ammonium salt-modified bentonite organoclay indeed offers additional flux aging loss enhancements via the epoxy system.

Three formulations were studied, demonstrating the effects of the additives on the overall properties. A standard coating formulation (STD) which consists only of the epoxy novolac resin, hardener, accelerator and lubricant is used as the benchmark. The two new formulations reported here (with respect to the STD samples) comprise:

1) an initial organotitanate precoat on the powders and subsequent coating of the novolac epoxy components and lubricant (NF1);
2) an initial organotitanate precoat on the powders and subsequent coating of the bisphenol A epoxy components, organoclay and lubricant (NF3).

Each component of the binder was mixed under high shear in the presence of an organic solvent and subsequently with the powders. Upon evaporation of the solvent, the powders were effectively coated.

Using a Powder Test Center (KZK Powder Tech. Corp.: PTC-03DT), the resultant coated powders were compacted into magnets with $\mathrm{PC} = 2$ (9.65 mm diameter × 6.35 mm height) at between 690 and 1250 MPa (7 and 12.8 Tcm^{-2}) which were then cured between 160 °C and 200 °C for between 30 and 60 min. Long term magnetic flux measurements were conducted at temperatures between 80 °C and 200 °C using fluxmeter and Helmholtz coil. Flux aging loss percentages were determined by dividing the change in flux by the initial flux. Hysteresigraph measurements on the $\mathrm{PC} = 2$ magnets yielded remanence and coercivity data. Tests performed on rectangular bars (28.73 mm × 9.77 mm × 2.93 mm) produced on a 200 kN (20-ton) press with an internally-designed die set determined three-point bending strengths, using a 450-kg (1000-lb) load cell and indicator to measure the maximum force applied to break each specimen.

III. RESULTS AND DISCUSSION

A. *Magnetic Properties*

Table I shows the effect of three different cure temperatures on the magnetic properties of the formulations compacted at 690 MPa (7 Tcm^{-2}). In general, the remanence and coercivity are reduced due to the increase in cure temperature. The NF formulations show higher coercivities than the STD formulation due to the presence of a more efficient powder coating. NF1 and NF3 have a reduced amount of magnetic powder due to the incorporation of the organic additives, thus their remanences are lowered. Powder A, however, shows a slightly improved $\mathrm{B_r}$ value for the NF3 formulation, due to its density being slightly larger than that of STD (see Fig. 1).

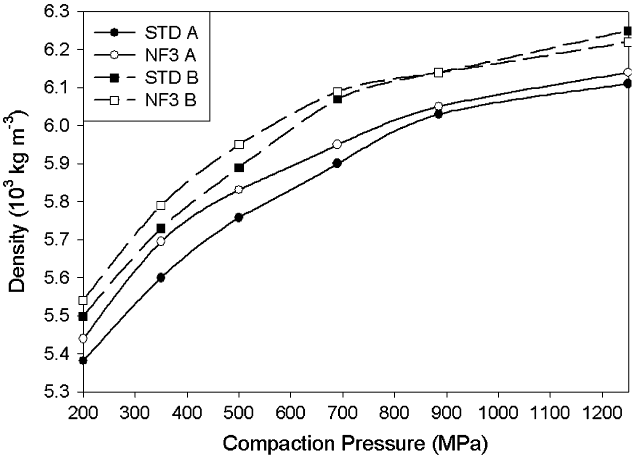

Fig. 1. Density as a function of compaction pressure for STD and NF3 formulations of both Powders A and B.

TABLE I
REMANENCE AND COERCIVITY VALUES FOR $\mathrm{PC} = 2$ MAGNETS COMPACTED AT 690 MPa AND CURED AT VARIOUS CURE TEMPERATURES FOR 60 MIN

	160°C		180°C		200°C	
	$\mathrm{B_r}$ mT	$\mathrm{H_{ci}}$ kA/m	$\mathrm{B_r}$ mT	$\mathrm{H_{ci}}$ kA/m	$\mathrm{B_r}$ mT	$\mathrm{H_{ci}}$ kA/m
STD A	703	749	702	739	701	725
NF1 A	697	760	696	758	696	756
NF3 A	704	758	705	756	706	754
STD B	684	923	681	922	680	918
NF1 B	679	940	679	942	677	938
NF3 B	681	942	676	937	679	937

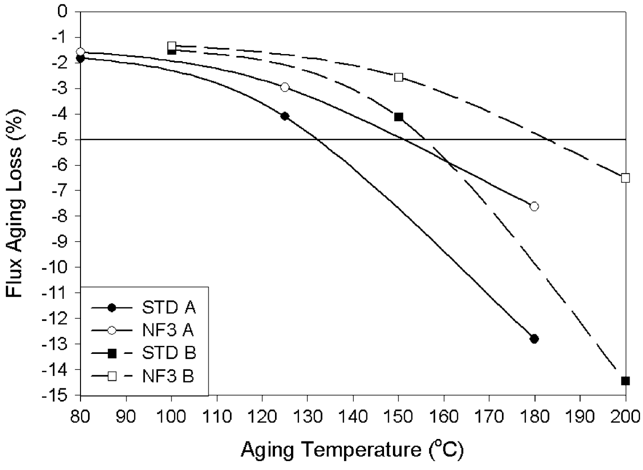

Fig. 2. Flux aging loss as a function of aging temperature of $\mathrm{PC} = 2$ magnets of two formulations of both Powder A and B compacted at 690 MPa, cured at 200 °C for 30 min and aged for 100 h.

B. *Flux Aging Loss*

All formulations for each powder type were tested, using aging temperatures of 80 °C–180 °C and 100 °C–200 °C for powder A and powder B magnets, respectively. Although all NF formulations showed improved flux aging loss behavior, only the best case (NF3) and worst case (STD) scenarios are included in Fig. 2. It can see seen from the curves in this figure that, for a given flux loss of −5%, in comparison to the STD A and B

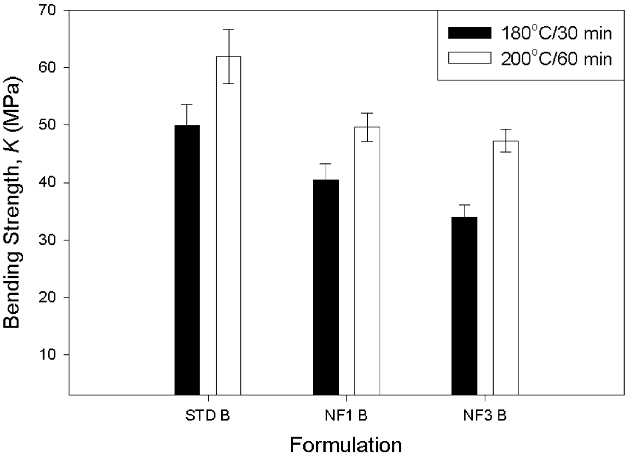

Fig. 3. Bending Strength values for three formulations of Powder B compacted at 690 MPa, cured at 180 °C for 30 min and 200 °C for 60 min.

magnets, temperature enhancements from 132 °C to 151 °C and from 156 °C to 182 °C, respectively, are gained when the NF3 formulation is incorporated. It was observed that for the highest compaction pressure (1250 MPa) and the lowest cure temperature (180 °C) the flux aging loss values were the worst, resulting in 9 °C–10 °C degradations of the operating temperatures for all of the magnets. The coated powder undergoes fragmentation and deformation under high pressure [4], revealing more exposed metal surfaces to the hot air. The high cure temperature of 200 °C allowed further cross-linking to occur within the compact, resulting in a protective coating with a better thermal stability and lower oxygen penetration than compacts cured at 180 °C.

C. Three-Point Bending Strength

Three-point bending strengths, K, were calculated from the load cell measurements using (1)

$$K = \frac{3IF}{2ht^2} \qquad (1)$$

I is the distance between the supports of the sample and equal to 15 mm for our calculations, F is the critical load required to break the sample, h is the sample height, and t is the sample thickness.

Fig. 3 represents the effect of cure conditions on the strength of the Powder B formulations. A high degree of cure of the epoxy components improves mechanical strength. With the NF formulations, the strength values decline with respect to the STD formulation. This can be attributed to the reduction in epoxy ingredients within the formulation due to the presence of the specialty additives and to the increase in cross-link density within the coating. Flexibility, which can be correlated to bending and tensile strengths, is known to behave inversely with cross-link density. The organotitanate has the ability to accelerate the epoxy polymerization reaction via catalysis by acting as a cross-linking agent [6]. Localized regions of high cross-link density in between the powder surface and the epoxy coating, where the organotitanate is precoated, may exist, giving rise to failure points in the cured magnets when subjected to a combination of tension and compression forces. However, compressive strengths (not shown) have been observed to increase when organotitanate is present within the coating.

For reasons not completely understood, the organoclay additive employed in NF3, although useful for oxidation resistance, may contribute to these strength reductions. First of all, the cross-link density may be increased due to the chemically-modified clay particles [5], thus, reducing the flexibility of the magnet. Secondly, our concentration of 10 wt% organoclay within the epoxy binder may be too high for good strength retention. Lastly, because of the extremely small size of the clay particles, it is possible that ineffective dispersion of them within the epoxy solution before coating may give rise to agglomerations on the powders, which represent regions within the coating where insufficient epoxy is present to form a strong bond between the powder particles.

IV. CONCLUSION

The NF formulations, which contain the precoat of an organotitanate adhesion agent and/or organoclay additive, offer excellent refinements to an organic coating used for liquid-coated RE Fe–B powders. They have proven to benefit magnet characteristics such as enhanced physical density, lower flux aging loss with little to no degradation in remanence and coercivity, with an acceptable reduction in bending strength. Bonded magnets produced from these formulations, cured at high temperatures (~180 °C–200 °C) and compacted at pressures around 690 MPa show favorable properties for use in applications at higher temperatures. For the two RE Fe–B powder types reported, these new formulations yielded between 19 °C and 26 °C enhancements for comparable flux aging loss.

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