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Annealing-Induced Property Improvements in 2-14-1 Powders Produced by Inert Gas Atomization

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Abstract—**The effects of vacuum annealing on the phase constitution and magnetic properties of various size fractions of 3 alloy compositions produced by inert-gas atomization (IGA) are examined. Annealing results in the oxidation of properitectic α-Fe formed during cooling of the melt, producing considerable improvement in the hard magnetic properties of the powders largely via the removal of lower-anisotropy magnetic reversal regions.**

I. Introduction

The Magnequench series of rapidly-solidified magnets based on the composition $RE_2Fe_{14}B$ is presently formed from the melt-spinning and subsequent annealing and crushing of ribbons of an arc-melted, rare-earth-rich precursor. As an alternative method in the production of rapidly-quenched powders for use in isotropic bonded magnets, inert gas atomization (IGA) offers the significant advantage over the conventional melt-quenching process that a large volume of material may be produced in a single step, using equipment that is more economical to operate than that presently used.

Previous work [1,2] has shown that while the IGA process is able to produce spherical, micron-sized Fe-Nd-B powders with high coercivities, the low energy products and powder nonuniformity are obstacles to further commercial development of the method. However, we have found that a brief vacuum annealing procedure raises both the coercivity and the remanence of the powder particle to levels comparable to those of typical bonded magnets. We present here the investigations of the physical changes responsible for the increase in the hysteretic magnetic properties with annealing, and offer suggestions for the further performance improvement of the particles.

II. Experimental

A laboratory-scale atomizer was used to produce powder in batches of 4 kg at a time. Master alloy ingots were made from commercial-grade powders and their composition was checked with ICP techniques. The nominal starting compositions are given in Table I; note that it proved to be necessary to enrich both the rare-earth and the boron concentration relative to that of the stoichiometric $Nd_2Fe_{14}B$ composition to obtain reasonable magnetic properties. The master alloys were then vacuum induction-heated in a high-purity Al_2O_3 crucible using a graphite susceptor before being atomized with helium. During atomization, the melt was superheated to 1400 °C to prevent freeze-off at the SiC nozzle.

The quenched powders were sieved in air to produce various size fractions, which are representative of different cooling rates and hence different microstructures. The powders were annealed in an infrared vacuum furnace at 650 °C for 10 minutes. The intrinsic coercivity H_{ci}, remanence B_R and saturation magnetization M_S were measured as a function of temperature using a Quantum Design MPMS SQUID magnetometer with a saturating field of 5.5 T and at room temperature using a VSM with a saturating field of 20.5 kG.

In both cases the powder samples were mixed with either stearic acid or wax and then consolidated into pellets to avoid individual particle rotation upon application of the applied field. No demagnetization corrections were applied. Powders were examined by x-ray diffraction performed at the National Synchrotron Light Source at Brookhaven National Laboratory using x-ray wavelengths that were chosen so as to avoid the excitation of Fe fluorescence. Lattice parameters were obtained using a least-squares-fit algorithm. Cursory examination of polished but unetched particles was performed with scanning electron microscopy (SEM).

III. Results

A. Magnetic Properties:

The magnetic properties of the alloys measured before and after annealing for selected particle size portions, Table II, show that the improvement in the energy product $(BH)_{max}$ upon annealing is largely due to a significant increase of the intrinsic coercivity; the remanence only shows increases on the order of 12 - 15%. The increase in coercivity with annealing is much larger for the larger-sized particle fraction. The increase of remanence that occurs with annealing must be due to the concurrent increase in coercivity, because in all cases the saturation magnetization measured at 20.5 kG exhibits either a decrease, or no significant change, with annealing. It should be noted that Batch 447 showed the least

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TABLE II
ROOM-TEMPERATURE MAGNETIC PROPERTIES OF IGA ALLOYS

IGA Alloy & Size Fraction	B_R (emu/g) Quenched	Annealed	H_{ci} (kOe) Quenched	Annealed	M_s (emu/g), @ 20.5 kG Quenched	Annealed	$(BH)_{max}$ (MGOe) Quenched	Annealed
Batch 434-2:								
10-20 μm	57.0	68.5	9.0	13.4	100.4	94.7	4.1	8.3
50-75 μm	47.0	63.9	2.6	8.9	94.6	93.0	2.0	6.8
Batch 435-2:								
10-20 μm	53.0	68.0	10	18	86.2	87.7	3.6	7.9
50-75 μm	54.0	63.9	4.2	13.7	85.0	85.0	3.6	7.5
Batch 447:								
10-20 μm	58.2	66.0	12.8	13.8	87.7	88.9	4.8	7.9
50-75 μm	58.5	66.6	5.4	12.8	91.1	90.0	4.4	8.0

McCallum [3] for the $Nd_2Fe_{14}B$ system is useful. A CCT diagram represents the expected constituent phases formed during solidification under continuous cooling conditions for a wide range of cooling rates. The resultant quench rates experienced by the particles are dependent upon such factors as the atomization gas used and the melt droplet size; typical atomization quench rates range from 10^4 to 10^5 K/sec [4]. From the arguments presented by Branagan and McCallum, in order to bypass the properitectic reaction solidification sequence that produces free iron, it is necessary to achieve a cooling rate such that the melt is undercooled below the 2-14-1 peritectic temperature. We are currently exploring the effects of carbide additives on the effective IGA quenching rate of alloys based on the $Nd_2Fe_{14}B$ composition [5].

V. CONCLUSIONS

Inert gas atomization performed on rare-earth-rich and boron-rich master alloys of the $RE_2Fe_{14}B$ composition produces powders with magnetic properties that improve upon annealing. The underlying phenomenon of this improvement is the oxidation of detrimental α-Fe which is formed during the properitectic reaction upon cooling of the melt. The α-Fe forms γ-Fe_2O_3, which causes a decrease in the saturation mag-netization M_S at 20.5 kG, but serves to improve both the remanence B_R and the coercivity H_{ci}. While this annealing-induced improvement provides powders with energy products $(BH)_{max}$ that exceed 8 MGOe for some powder size fractions, the powder uniformity and remanence are still rather poor. To improve the magnetic properties of 2-14-1-based powders produced by IGA, efforts are underway to increase the effective quench rate experienced by the melt via carbide additions to the master ingot [5].

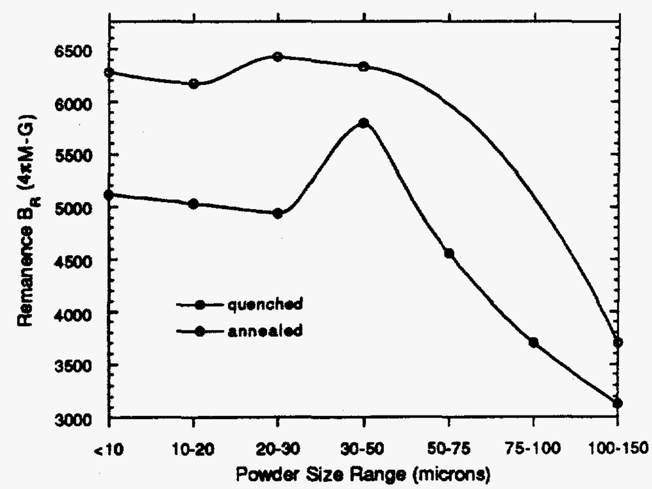

Fig. 3. The remanence B_R as a function of powder size fraction for as-quenched and annealed powders from Batch 435-2.

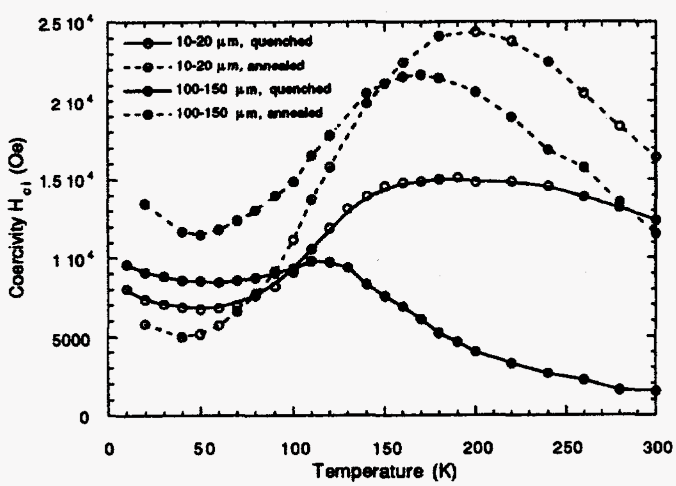

Fig. 2. Coercivity H_{ci} as a function of temperature for as-quenched and the annealed samples from the 10-20 μm and the 100-150 μm powder size fractions of Batch 435-2.

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