Water milling and gas passivation method for production of corrosion resistant Nd FeBN/C powder and magnets

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Water milling and gas passivation method for production of corrosion resistant Nd-Fe-B-N/C powder and magnets

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Nd-Fe-B powder produced by conventional methods is pyrophoric, and exhibits poor corrosion resistance. Magnets made from powder are also susceptible to corrosion. Conventional methods of production are complicated, potentially hazardous, and relatively expensive. A novel, low cost, less hazardous method of producing powder and magnets with high corrosion resistance and Curie point is discussed. Nd-Fe-B alloys are milled in water, vacuum dried, and passivated at a suitable temperature in a nitrogen or carbon dioxide medium. During passivation, a protective layer, comprised of nitride and/or carbide phases, is formed in the surface region of the powder particles. This powder is not pyrophoric, and may be stored in a laboratory air environment for an extended period of time with no loss in magnetic properties. Compacted and sintered magnets produced from this powder are also highly corrosion resistant, and exhibit a higher Curie point compared to conventionally produced magnets. A description of the novel technology, and a discussion of the properties of Nd-Fe-B-C/N powder and magnets is given.

INTRODUCTION

The majority of rare-earth magnets are produced by powder metallurgical techniques. Cast ingots of the alloy are broken down to a coarse powder. This powder is then milled in an attritor or jet mill to break it down into single crystal particles, with one easy magnetization axis.

In attritor milling, a protective environment is necessary as Nd-Fe-B powder is pyrophoric, and will easily oxidize. Organic liquids such as toluene, methanol, petroleum ether, or hexane are used because they allow a hydride layer to form on the particle surfaces, lessening the rate at which the powder will oxidize. However, unless the powder is compacted as soon as possible after milling, oxidation will occur. The powder is compacted in a magnetic field to orient the particles, then sintered, heat treated, and machined.

This method of manufacture gives rise to several problems. The liquids used are expensive, toxic, and flammable, and therefore present potential hazards of fire and explosion. Because of this, all equipment and facilities used in the process must be fire and explosion proof. In addition, the powder and powder compacts must be handled in a protective atmosphere, adding to the cost of the equipment and process.

EXPERIMENTAL METHODS

Ingots of Nd-Fe-B alloy were vacuum induction melted and cooled in a copper mold. The ingots were crushed in a disk pulverizer to 50 mesh size powders. This powder was composed of two lots, one of 34% Nd, and one of 39.1% Nd content. The powders were mixed together, in calculated ratios, to produce several lots with Nd contents of 34% to 39.1%. In addition, various amounts of dysprosium were added to several lots of the powder. The powders were processed as illustrated in Fig. 1:

(1) Samples were milled in an attritor, in water with various powder/ball ratios and times.
(2) The wet powders were vacuum dried at 40–90 °C, in a flow of neutral gas at negative pressures, for 1–4 h.
(3) The dried powders were passivated at 125–300 °C in carbon dioxide and/or nitrogen gas.
(4) The passivated powders were compacted, in a laboratory air atmosphere, at 3 to 8 T/cm^2 in a magnetic field.
(5) The powder compacts were sintered at 1060–1125 °C, for 1–4 h, and heat treated with multistage heat treat cycles at 400 to 1000 °C.

The susceptibility of the powder and magnet samples to corrosion was determined by measuring weight gain and

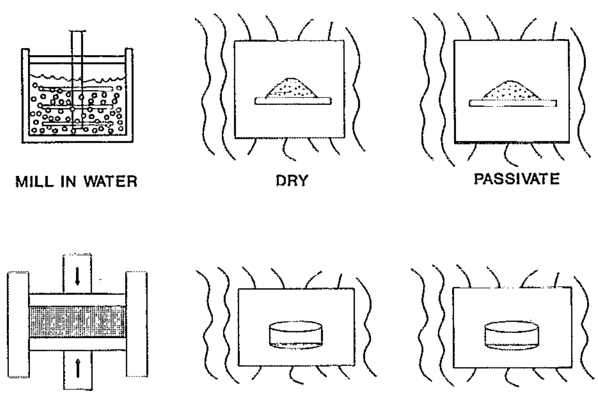

FIG. 1. Schematic of water milling and gas passivation process for the production of sintered magnets.

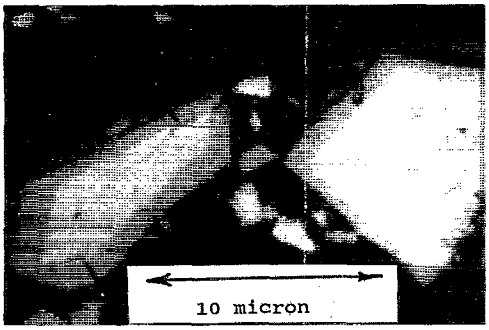

FIG. 2. Photomicrograph of water milled and gas passivated powder particles.

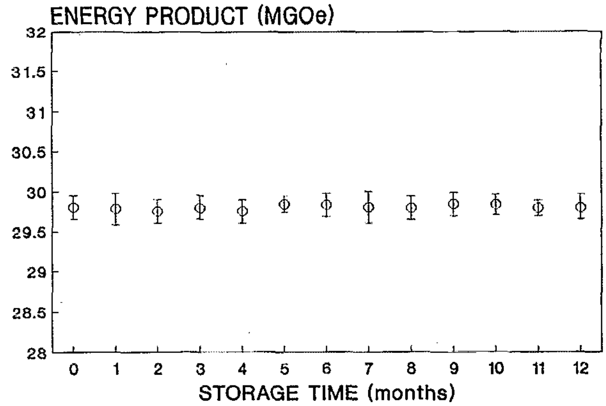

FIG. 4. Maximum energy product of magnets made from water milled and gas passivated powder which had been stored in laboratory air environment.

magnetic properties after exposure to laboratory air and high relative humidity environments for various times and temperatures.

Magnetic properties and Curie temperature of the sintered magnets and powders were measured on a vibrating-sample magnetometer (VSM) and automatic magnetic hysteresisgraph. Chemical composition and phase analysis were evaluated by standard Auger spectroscopy and x-ray diffraction (XRD) methods.

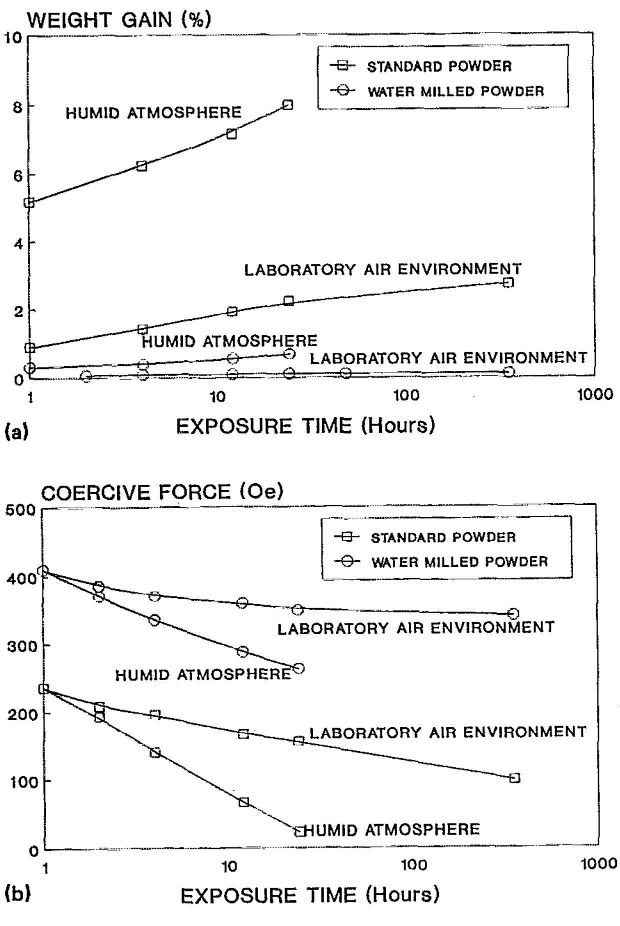

FIG. 3. (a) Weight gain of powders during exposure to humid and laboratory air environments and (b) coercivity losses in powders during exposure to humid and laboratory air environments.

RESULTS AND DISCUSSION

The magnetic properties of water milled and gas passivated powder are influenced by milling and passivating parameters, such as time and temperature. Passivation of the powder results in absorption of nitrogen or carbon and subsequent diffusion of these atoms within the crystal lattice of the surface layers (Fig. 2). The passivated layer contained $Nd_2Fe_{14}B$, and Nd_2C_3 and NdC_2 phases which protect the particles from corrosion.

This powder, after corrosion tests, exhibited a much lower weight gain, and retained higher magnetic properties, compared to conventionally produced powders (Fig. 3). After exposure, small amounts of NdO and Nd_2O_3 phases were observed on powder surfaces.

Water milled and gas passivated powders may be stored in a laboratory environment for extended periods with no loss of magnetic properties. Magnets compacted and sintered up to six months after milling (Fig. 4) exhibited no appreciable loss in magnetic properties when compared to magnets compacted immediately after milling of the powder.

The magnetic properties of sintered magnets are affected by milling time, passivation temperature and time,

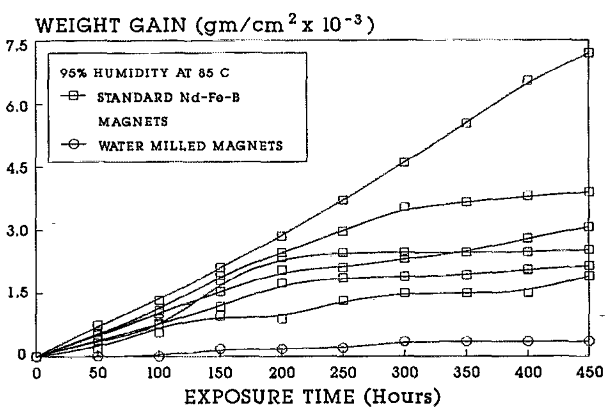

FIG. 5. Weight gain of Nd-Fe-B magnets made from water milled and gas passivated powder during exposure to humid atmosphere.

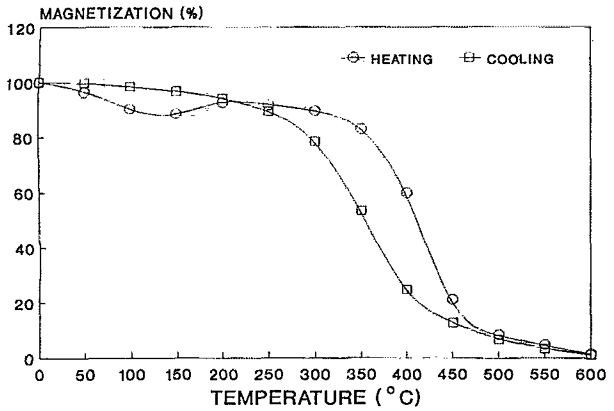

FIG. 6. Magnetization vs temperature of Nd-Fe-B-C magnets produced from water milled and gas passivated powder.

compacting pressure, and sintering temperature and time. The highest magnetic properties of as-sintered magnets are $(BH)_{\max} = 29.0$ MGOe, $B_r = 11.8$ kG, and ${}_iH_c = 10.5$, achieved after processing under optimum conditions.

Maximum energy product and coercive force of the sintered magnets are also affected by heat treatment parameters, such as temperature, time, cooling rate, and process sequence. Maximum properties $(BH)_{\max} = 33.43$ MGOe and ${}_iH_c = 14.13$ kOe were achieved in heat treated magnets.

After exposure to conditions of high relative humidity at 85 °C, magnets made from the water milled and gas passivated powders exhibited a much lower weight gain than that exhibited by conventionally processed magnets (Fig. 5) obtained from several major industry suppliers. This indicates that the magnets produced from water milled and gas passivated powder retained the protective carbide/nitride containing layer on grain surfaces.

The existence of this layer also improved thermal stability and Curie temperature. The Curie point for the Nd-Fe-B-N/C magnets is estimated to be approximately 440 °C (Fig. 6), well above the usually reported value of 310 °C for conventionally processed magnets.

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