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d-HDDR Processing of Nd-Fe-B Based Alloys to Obtain Highly Anisotropic Nanocrystalline Powders

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Abstract. The HDDR (hydrogenation-disproportionation-desorption-recombination) process is an established powder metallurgy route to obtain Nd–Fe–B nanocrystalline powders for bonded magnets manufacturing. Therefore, both conventional (c-HDDR) and dynamic HDDR (d-HDDR) processes has been investigated to obtain Nd-Fe-B-based powders with different characteristics. Magnetic properties were measured by means of a hysteresisgraph and the powder obtained by d-HDDR showed strong anisotropy, allowing a B_r of 1.1 T in the bonded magnet, whereas c-HDDR powder was isotropic with a B_r of 0.6 T. Microstructural changes were characterized by X-ray diffraction (DRX) and scanning electron microscopy (SEM). X-ray patters of anisotropic powders made by d-HDDR showed high intensity reflection peaks indexed as (004), (105) and (006) planes in the aligning direction, due the texture inducement in c-axis of the main phase ($Nd_2Fe_{14}B$). However, SEM micrographs of c-HDDR powder showed a more homogeneous microstructure, with grain size of ~300 nm, when compared to d-HDDR powder that ranged from 300 nm to 500 nm. This difference is assumed to be the cause of lower intrinsic coercivity found in the c-HDDR powder.

Introduction

Currently, the demand for high performance permanent magnets in green technology applications, such as wind turbines and hybrid electric vehicles, has been increasing every year. Therefore, research topics on Nd-Fe-B anisotropic powder for bonded magnet fabrication has been conducted using a well-known thermo-chemical technique called hydrogenation-disproportionation-desorption-recombination (HDDR) [1,2]. In this process, hydrogen is absorbed at a high temperature (~750–900 °C) by $Nd_2Fe_{14}B$ phase, which causes its disproportionation into three phases: NdH_2; α-Fe and Fe_2B. After this step, the desorption of the hydrogen induces the recombination of the three phases recombine into a fine-grained $Nd_2Fe_{14}B$ structure, with grain size in the range of 300 nm [3]. Although the conventional HDDR (c-HDDR) is effective for obtaining hard magnetic materials, anisotropic powders cannot be obtained by this method. Thus, many studies have been carried out to understand the mechanism of the texture development in HDDR powders, such as the addition of elements Co, Ga, Nb and Zr are added to the initial alloy [3,4]. Other authors reported that by maintaining the ideal temperature, hydrogen pressure and reaction speed, anisotropy can be achieved by the texture memory effect (TME) between $Nd_2Fe_{14}B$ and Fe_2B phases (d-HDDR phenomenon, where d=dynamic) [4,5]. In this work, d-HDDR was compared with c-HDDR, showing the potential of d-HDDR as a promising technique to obtain highly Nd-Fe-B anisotropic powders.

Experimental

Conventional and dynamic HDDR cycles were performed using as starting material a commercial N42 grade Nd-Fe-B magnet. Prior to HDDR treatments, the magnet was subjected to the fragilization process by hydrogen decriptation, which generates a coarse powder that serves as feeding material for HDDR treatments. Thereafter, c-HDDR cycle was carried out under 100 kPa hydrogen pressure at 1123 K for 10 minutes for disproportionation. After full disproportionation, recombination was performed at the same temperature for 15 minutes by the application of a vacuum below 1 Pa, followed by rapid cooling. In contrast, d-HDDR cycle was carried out under 30 kPa hydrogen pressure at 1093 K for 180 minutes for controlled disproportionation. After this step, recombination was performed at the same temperature, but in two steps, first inducing a partial recombination for 15 minutes under 10 kPa, and then a final recombination step for 15 minutes below 1 Pa, also followed by rapid cooling. The HDDR processed materials were then milled and mixed with an epoxy resin into a cubic mold subjected to a constant magnetic field for particle alignment. In order to evaluate the magnetic properties, HDDR samples were magnetically saturated under a pulsed magnetic field with 4,5 T intensity and, subsequently, characterized by means of a hysteresisgraph. Further, results were normalized by the volume ratio between powder and epoxy resin in order to obtain magnetic properties of HDDR powders. Microstructural aspects were investigated by X-ray diffraction (XRD) with Cu-K_α radiation and by scanning electron microscopy (SEM).

Results and Discussion

Fig. 1 shows the demagnetization curve of the powders obtained by c-HDDR and d-HDDR, which shows d-HDDR as an effective route for producing anisotropic powders with B_r of ~1.1 T, $(BH)_{max}$ of 83 kJ/m³, and high anisotropy level. On the other hand, it was found that optimum values for intrinsic coercivity were achieved using c-HDDR route, with a H_{cj} of ~830 kA/m, whereas for d-HDDR powders, the intrinsic coercivity measured in the easy axis was ~700 kA/m.

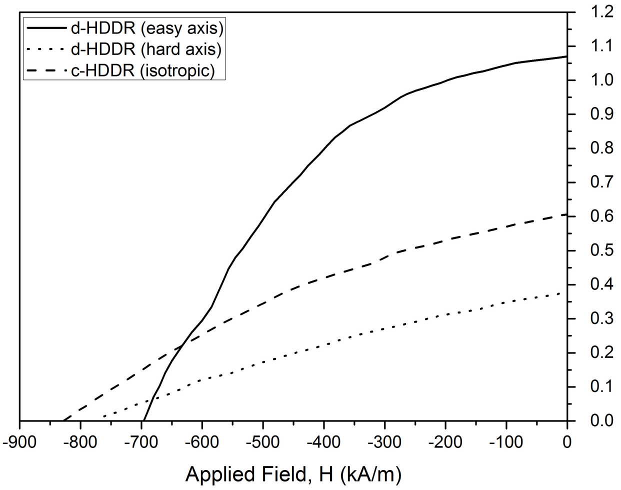

Fig. 1: Demagnetization curve of c-HDDR and d-HDDR powders

The high level of texture of d-HDDR bonded samples can also be demonstrated by XRD analysis. Fig. 2 shows the results of XRD patterns obtained in the alignment direction (easy axis) of d-HDDR bonded magnets, as well as in the hard axis (parallel to alignment) of the same sample and the XRD pattern of the c-HDDR bonded magnet (which is similar for every direction). As can be seen in the X-ray analysis for the easy axis of d-HDDR magnet, it shows a strong reflection in the angles indexed as planes (004), (105) and (006) of the $Nd_2Fe_{14}B$ phase, a consequence of the alignment in <00l> crystal direction of d-HDDR textured particles during field curing process. As expected, these preferential reflections were not found in the c-HDDR bonded magnet X-ray analysis, due to its isotropic condition.

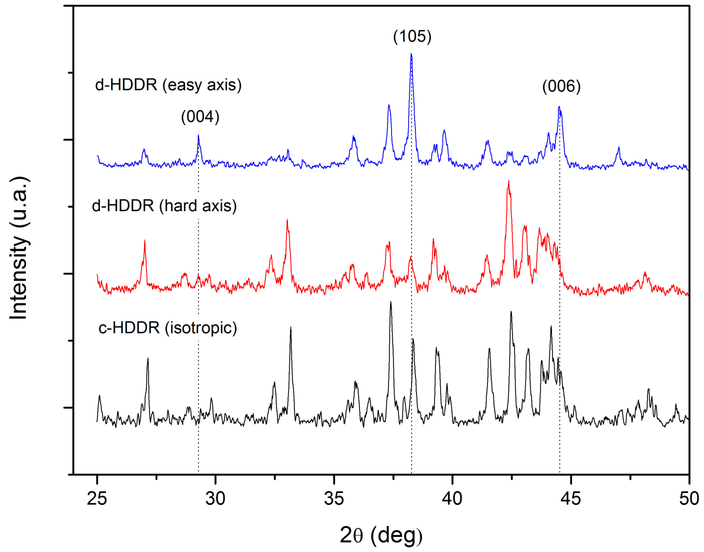

Fig. 2: X-ray patterns of d-HDDR (easy and hard axis) and c-HDDR bonded magnets

Fig. 3 and Fig. 4 show SEM micrographs of $Nd_2Fe_{14}B$ recombined grains for c-HDDR and d-HDDR powders, respectively. Statistical analysis showed an average grain size for the c-HDDR powder around 300 nm, with a more homogeneous distribution compared to the d-HDDR powder. It can be observed that the average grain size of d-HDDR powder is also approximately 300 nm, however, there are larger grains of up to 400 nm. The fact that larger grains are present in the d-HDDR powder could also be a reason for the lower intrinsic coercivity of this material, since this property is strongly dependent on the grain size in the case of Nd-Fe-B-based alloys processed by HDDR [5,6]. Once the single domain size for $Nd_2Fe_{14}B$ phase is around 250 nm, HDDR powders with grains beyond this critical size may induce lower intrinsic coercivity [6].

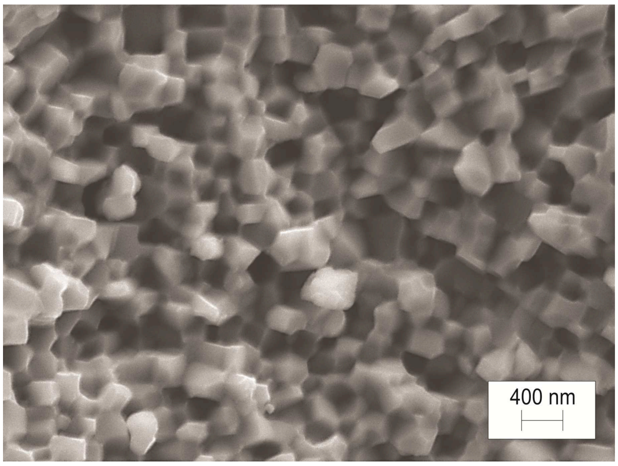

Fig. 3: SEM image of c-HDDR powder

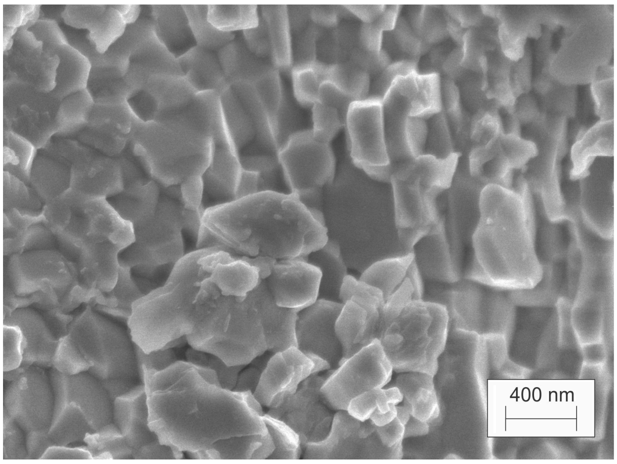

Fig. 4: SEM micrograph of d-HDDR powder

Conclusions

Present results demonstrate that the d-HDDR process is an effective method to enhance the magnetic properties of Nd-Fe-B based-powders for bonded magnets by the development of crystallographic texture. The results showed that B_r and H_{cj} values are affected by controlled disproportionation by hydrogen pressure (~30 kPa), which induces crystallographic texture in nanocrystalline Nd-Fe-B powders. X-ray analysis of c-HDDR and d-HDDR in field-cured samples confirmed the presence of texture in the d-HDDR processed powder, whereas c-HDDR was found to be isotropic, a consequence of random grain orientation resulted by the lack of pressure control in the disproportionation step.

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