

# Thermal Stability of Hddr-treated Nd-Fe-B-type Material

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## 1. Introduction

The hydrogenation, disproportionation, desorption and recombination (HDDR) process has been established as an effective technique for producing highly coercive fine Nd-Fe-B magnetic powder. As the HDDR-treated material consists of very fine grains with size comparable to single domain size (~300 nm for Nd<sub>2</sub>Fe<sub>14</sub>B), it would be expected that consolidation of the HDDR-treated powder could lead to bulk magnet with fine grain structure, thus leading to high coercivity. An attempt was made in our laboratory to consolidate the HDDR powder by hot-pressing. However, coercivity of the hot-pressed compact was radically reduced above 650 °C, and this was attributed to the residual hydrogen[1]. Due to the residual hydrogen, the HDDR-treated powder lost radically its coercivity when heated up to an elevated temperature. The HDDR-treated powder exhibits a poor thermal stability unless the residual hydrogen is completely removed before thermal processing. In this article, thermal stability of the HDDR-treated powder was studied in detail, and it was correlated with the residual hydrogen in the powder.

## 2. Experimental works

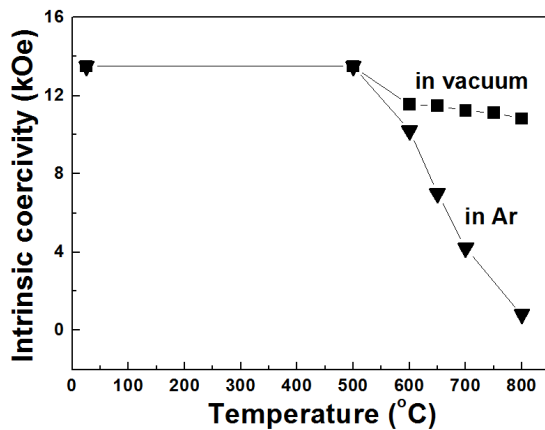
The starting material used in the present study is HDDR-treated powder of Nd<sub>12.5</sub>Fe<sub>80.6</sub>B<sub>6.4</sub>Ga<sub>0.3</sub>Nb<sub>0.2</sub> alloy (iHc =13.5 kOe). The thermal stability of HDDR-treated powder was studied over a temperature range from 500 °C to 800 °C in vacuum and Ar atmospheres with heating rate of 25 °C/min. Magnetic characterization of the materials was undertaken by means of vibrating sample magnetometer (VSM) with a maximum field of 12 kOe. X-ray diffractometer (XRD) (Cu-K $\alpha$  radiation) and differential thermal analysis (DTA) were used for studying the phase change in the HDDR-treated powder during heating. The HDDR powder pre-degassed by heating up to 800 °C under vacuum was re-heated over a temperature range from 600 °C to 800 °C in Ar and vacuum.

## 3. Results and discussion

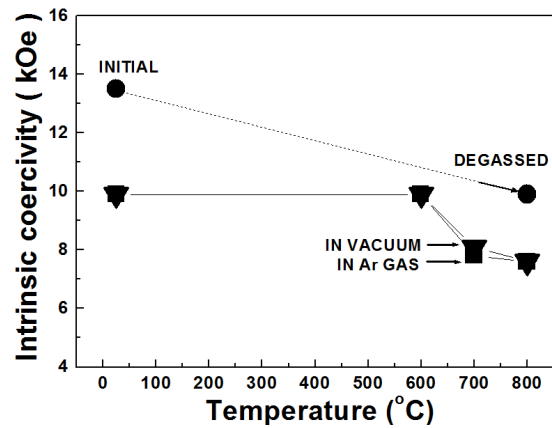
The Nd<sub>12.5</sub>Fe<sub>80.6</sub> B<sub>6.4</sub>Ga<sub>0.3</sub>Nb<sub>0.2</sub> HDDR-treated powder was heated in vacuum and Ar atmosphere over the temperature ranges from 500 °C to 800°C. The coercivity of the heated specimens was measured, and results are shown in Fig. 1. It appears that coercivity of the powder was reduced significantly above 600°C, and interestingly, the coercivity reduction was much more profound for the material heated in Ar atmosphere than in vacuum. One of the possible reasons for the coercivity reduction would be an excessive grain growth caused by the heating at elevated temperature. However, this is not the case proved by SEM micrographs. Our earlier work revealed that the HDDR-treated powder contained significant amount of hydrogen (approx. 1500 ppm)[1]. The residual hydrogen was considered to exist in the form of Nd<sub>2</sub>Fe<sub>14</sub>BH<sub>x</sub> hydride, and it was disproportionated into  $\alpha$ -Fe, Fe<sub>2</sub>B and NdH<sub>2</sub> phases. The magnetically soft  $\alpha$ -Fe and Fe<sub>2</sub>B may be responsible for the radical coercivity reduction above 600°C in Ar atmosphere. The residual hydrogen may be desorbed more effectively in the course of heating in vacuum than in Ar gas atmosphere. This may explain the fact that coercivity reduction is less profound for the sample heated in vacuum. Disproportionation of the Nd<sub>2</sub>Fe<sub>14</sub>BH<sub>x</sub> hydride in the sample

heated in Ar was verified by XRD phase analysis. Patterns were almost similar except that  $\text{Fe}_{2\text{B}}$  and  $\alpha\text{-Fe}$  phases existed along with  $\text{NdH}_2$  in the powder heated in Ar formed via disproportionation of the  $\text{Nd}_2\text{Fe}_{14}\text{BH}_x$  in the HDDR powder, leading to radical coercivity reduction above  $600^\circ\text{C}$ . DTA results also confirmed the disproportionation of the  $\text{Nd}_2\text{Fe}_{14}\text{BH}_x$  hydride in the sample heated in Ar at the temperature range of  $620^\circ\text{C} - 655^\circ\text{C}$ .

The present study suggests that poor thermal stability of the HDDR-treated powder is due largely to the residual hydrogen. Then, it would be expected that the HDDR-treated powder may show similar coercivity variation during heating regardless of the atmosphere if the residual hydrogen is degassed previously. In order to see whether the previously degassed powder show similar coercivity variation during heating regardless of the atmosphere, the degassed powder (by heating up to  $800^\circ\text{C}$  under vacuum) was heated in Ar and vacuum, and the coercivity variations were shown in Fig. 2. Also included in Fig. 2 are the coercivity values in initial powder and in degassed state for comparison. It can be seen that coercivity variation of the previously degassed material in the course of heating is almost identical regardless of the atmosphere Ar or vacuum. Considering the radical coercivity reduction in the initial powder heated in Ar atmosphere (Fig. 1), the present finding that coercivity variation of the previously degassed material is almost identical regardless of the atmosphere is somewhat striking. It can be said, therefore, that the poor thermal stability of the HDDR-treated powder in terms of coercivity can be attributed almost certainly to the residual hydrogen.



**Fig. 1.** Coercivity variation of  $\text{Nd}_{12.5}\text{Fe}_{80.6}$   $\text{B}_{6.4}\text{Ga}_{0.3}\text{Nb}_{0.2}$  HDDR-treated powder heated under vacuum or Ar atmosphere.



**Fig. 2.** Coercivity variations of the  $\text{Nd}_{12.5}\text{Fe}_{80.6}$   $\text{B}_{6.4}\text{Ga}_{0.3}\text{Nb}_{0.2}$  HDDR-treated powder previously degassed by heating up to  $800^\circ\text{C}$  under vacuum.

## Reference

- [1] M. A. Matin, H. W. Kwon, J. G. Lee, J. H. Yu, T. H. Kim and C. W. Yang, IEEE Transaction on Magnetics, (2013), in press.