Anisotropy inducement mechanism in hydrogen disproportionation desorption recombination (HDDR) processed Nd-Fe-B powders

H. Sepehri-Amin¹, T. Ohkubo¹, K. Hono¹, K. Güth², and O. Gutfleisch²,³
¹Elements Strategy Initiative Center for Magnetic Materials, NIMS, Tsukuba, Japan
²Fraunhofer ISC Projektgruppe IWKS, Germany
³Materialwissenschaft, Technische Universität Darmstadt, Germany

Introduction

The hydrogenation-disproportionation-desorption-recombination (HDDR) process is an attractive and unique method for producing anisotropic nanocrystalline Nd-Fe-B powders. In order to develop highly textured Nd-Fe-B powders, HDDR process parameters need to be carefully chosen and controlled [1]. Although lots of investigations have been carried out to understand the mechanism of the anisotropy development in HDDR powders [1-3], some questions still remain. In this work, the microstructures of Nd-Fe-B powders that were HD processed at different hydrogen pressure ($P_{\text{H}_2}^{\text{HD}}$) were investigated to fully clarify the mechanism of the anisotropic microstructure evolution.

Experimental

Dynamic hydrogenation disproportionation (HD) desorption recombination (DR) process was carried out on Nd$_{12.8}$Fe$_{80.1}$B$_{6.6}$Ga$_{0.3}$Nb$_{0.2}$ alloy powders at different HD hydrogen pressure, $P_{\text{H}_2}^{\text{HD}} = 30$ kPa and 100 kPa. The microstructures of the samples in early stages of the HD process, a fully HD process, and an early DR process were investigated using SEM/FIB (Carl Zeiss 1540EsB), TEM (Titan G2 80-200), and a locally built laser-assisted three dimensional atom probe (3DAP) to characterize the memory sites responsible for the texture development.

Results

A high remanent magnetization of 1.43 T was obtained for the fully HDDR processed powder with $P_{\text{H}_2}^{\text{HD}} = 30$ kPa, indicating a strong [001] crystallographic texture. However, $P_{\text{H}_2}^{\text{HD}} = 100$ kPa led to weakly textured Nd-Fe-B powders with a remanent magnetization of 0.89 T.

TEM observations from early HD processed powder with $P_{\text{H}_2}^{\text{HD}} = 30$ kPa showed that the Fe$_2$B phase has a direct crystallographic orientation relationship with the initial Nd$_2$Fe$_{14}$B grains, i.e. [420]$_{\text{Fe}_2\text{B}}$ || [211]$_{\text{Nd}_2\text{Fe}_{14}\text{B}}$ and (002)$_{\text{Fe}_2\text{B}}$ || (110)$_{\text{Nd}_2\text{Fe}_{14}\text{B}}$. Energy filtered (EF)-TEM and 3DAP results obtained from a fully HD processed sample showed boron not only in the Fe$_2$B phase but also at the NdH$_2$/Nd$_2$-Fe interfaces in both weakly and highly textured samples. High resolution STEM-HAADF image and nano-beam diffraction analysis from NdH$_2$/α-Fe interfaces showed that boron enrichment at these interfaces does not make a separate phase, such as iron boride. However, there is boron enrichment more in Fe grain of the NdH$_2$/α-Fe interface in nano-scale. Fig. 1(a) shows EF-TEM maps of B and Nd for the fully HD processed sample with $P_{\text{H}_2}^{\text{HD}} = 30$ kPa. Bright field (BF)-TEM images obtained from the fully HD processed samples with different $P_{\text{H}_2}^{\text{HD}}$ showed that the Fe$_2$B regions in both
weakly and highly textured samples comprise of several small Fe$_2$B grains. Fig. 1 (b) and (c) shows BF and dark field (DF)-TEM images obtained from fully HD processed P$_{HD}$=30 kPa sample. The DF-TEM image shows that the Fe$_2$B sub-grains in boride region of fully HD processed P$_{HD}$ = 30 kPa sample is strongly textured. Fig. 2 (a) shows BF-TEM image taken from the sample fully HD processed at P$_{HD}$ = 100 kPa. Superimposed EF-TEM maps of Nd (red), Fe (Blue), and B (green), obtained from the same region as Fig. 2(a), is shown in Fig. 2 (b). By comparing Fig. 2 (a) and (b), NdH$_2$, α-Fe, and Fe$_2$B phases can be distinguished. Selected area diffraction patterns obtained from different Fe$_2$B grains in boride area are shown in Fig. 2(c), indicating that the Fe$_2$B grains in the boride area are not well aligned in the sample fully HD processed at P$_{HD}$ = 100 kPa. Orientation relationship study of P$_{HD}$ = 30 kPa sample at early DR processed stage showed that recombined Nd$_2$Fe$_{14}$B grains have direct orientation relationship with the remaining Fe$_2$B phase from HD process. In addition, 3D SEM tomography obtained from 3D serial sectioning of BSE images from P$_{HD}$ = 30 kPa sample at very early stage of DR process showed that the recombined Nd$_2$Fe$_{14}$B grains nucleate at the interface of Fe$_2$B/NdH$_2$ grains and grow through the interface of NdH$_2$/α-Fe grains.

These microstructure investigations indicate that the highly aligned Fe$_2$B grains act as memory sites for the development of the texture in the sample HD processed at P$_{HD}$ = 30 kPa, as shown schematically in Fig. 3, consistent with the previously proposed texture memory effect (TME) model. Importantly, it can now be shown that the recombined Nd$_2$Fe$_{14}$B phase nucleates at the interface of Fe$_2$B with NdH$_2$ phase and grow through the interface of NdH$_2$/α-Fe interfaces and boron segregated at the NdH$_2$/α-Fe interface acts as a boron source for the growth of the recombined Nd$_2$Fe$_{14}$B grains during DR process.

Reference