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EFFECTS OF WATER QUENCHING BEFORE HYDROGENATION, DISPROPORTIONATION, DESORPTION, AND RECOMBINATION PROCESS ON THE MAGNETIC PROPERTIES OF Nd-Fe-B POWDER

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ABSTRACT

In general, the Nd-Fe-B as-cast ingot is homogenized for a long time before the hydrogenation, disproportionation, desorption and recombination (HDDR) process, to produce good magnetic properties. Since the homogenizing process is expensive, this work examined the possibility of replacing it with a water quenching process for Nd-Fe-B magnetic powder. The magnetic powders were soaked at 1100^{o}C for 4 hours, followed by water quenching prior to HDDR. The resulting powder had magnetic properties that were almost similar to magnetic powder that was homogenized prior to HDDR. The remanence values of the water-quenched alloy and the homogenized alloy were 7.8 KG and 8.9 KG, respectively, while the coercivity values were 12.6 KOe and 10.3 KOe, respectively. In general, the magnetic properties of both samples were not much different. The microstructure of $Nd_2Fe_{14}B$ phase combined with a very slight Nd rich phase in micro grain boundary likely caused the coercivity enhancement.

Keywords: Coercivity; Homogeneizing; HDDR; Water quenching

1. INTRODUCTION

The Research on the high performance $Nd_2Fe_{14}B$ based permanent magnet has recently increased, in accordance with the extensive development of hybrid and electric cars. The room temperature coercivity of the commercially available $Nd_2Fe_{14}B$-based permanent magnets is relatively low, so it is not suitable for high temperature operation in the traction motors of hybrid and electric cars. One method for achieving high coercivity is refining the grain size of the $Nd_2Fe_{14}B$ phase. The hydrogenation, disproportionation, desorption, and recombination (HDDR) process is an effective and economic way for refining the grain of the $Nd_2Fe_{14}B$ phase and inducing anisotropic magnet properties in $Nd_2Fe_{14}B$ alloys (Ahmed et al., 2008; Sepehri-Amin et al., 2010; Faria et al., 2000; Kawashita et al., 2004). These HDDR processed powders have the ultrafine grain of the $Nd_2Fe_{14}B$ phase.

$$Nd_2Fe_{14}B + 2H_2 \longrightarrow 2NdH_2 + 12\ Fe + Fe_2B \quad \text{(HD process)}$$
$$2NdH_2 + 12Fe + Fe_2B \longrightarrow Nd_2Fe_{14}B + 2H_2 \quad \text{(DR process)}$$

The HDDR process composes the hydrogenation-disproportionation (HD) process and the desorption-recombination (DR) process, as shown below. Prior to the HDDR process, the as-

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cast ingot is generally homogenized for a long time in order to obtain the single phase of a homogenized structure (Ahmed et al., 2008; Faria et al., 2000; Kawashita et al., 2004). This homogenization process requires additional energy and thus additional costs. As previously reported, the HDDR magnetic powders prepared directly from the as-cast ingots exhibit lower magnetic properties than those prepared from homogenized as-cast ingots, due to the existence of soft magnetic α-Fe in the as-cast ingots (Cannesan et al., 2001; Faria et al., 2000; Kawashita et al., 2004).

Rapid cooling is one way to prevent the segregated phase in order to produce a single phase. One rapid cooling method is water quenching. In order to reduce the time and offer a simple process, this work examined the possibility of using water quenching to replace the homogenizing of the magnetic powder. Accordingly, this paper discusses the effects of using different soaking temperatures before water quenching on the magnetic properties of HDDR-processed Nd-Fe-B powder.

2. EXPERIMENTAL SETUP

20 gram samples of as-casted alloys with a chemical composition of $Nd_{12.5}Fe_{80.6}B_{6.4}G_{0.3}Nb_{0.2}$ were prepared by an argon atmosphere arc melting furnace. The as-casted alloys were carried out in an evacuated quartz tube by soaking the alloys at different high temperatures for 4 hours with argon flow, and then cooling them in water. Then the quenched alloys were crushed to small granules and the HDDR process was performed. Figure 1 shows the conditions used for the HDDR process. Pure 100% H_2 was used in this experiment.

The HDDR-processed powders were crushed to a particle size of less than 150 µm, mixed with paraffin, and were measured using a vibrating sample magnetometer (VSM) after magnetic alignment, using a static magnetic field with a strength of 800 kA m^{-1}, and then magnetized using a maximum magnetic field pulse of 2 T. The alloy microstructures before and after HDDR processing were observed using a scanning electron microscope - back scatter electron (SEM-BSE) image and energy dispersive spectroscopy (EDS) analysis.

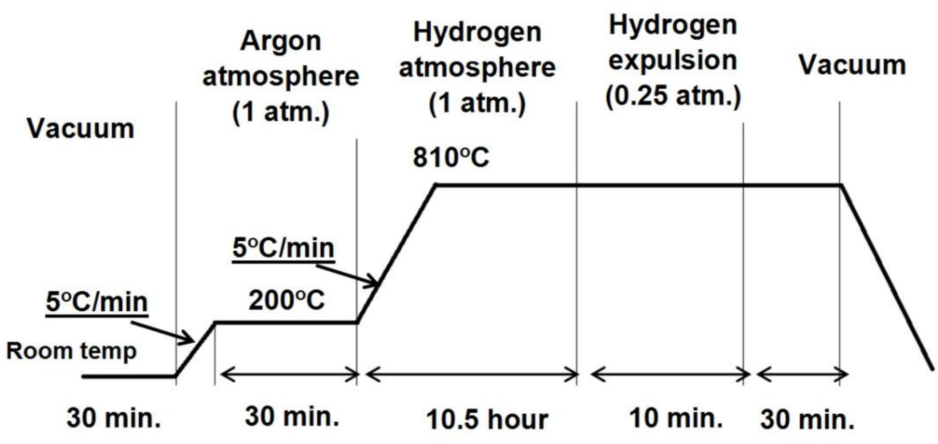

Figure 1 The HDDR process.

3. RESULTS

Figure 2 shows the SEM-BSE image of an Nd-Fe-B as-casted alloy. As reported in previous investigations, Nd-Fe-B as-casted alloys usually have microstructures characterized by a combination of large amounts of α-Fe dendrites (the dark region), $Nd_2Fe_{14}B$ phase (the gray region), and some Nd-rich phases (the white region) (Liu et al., 2009; Cannesan et al., 2001; Kawashita et al., 2004).

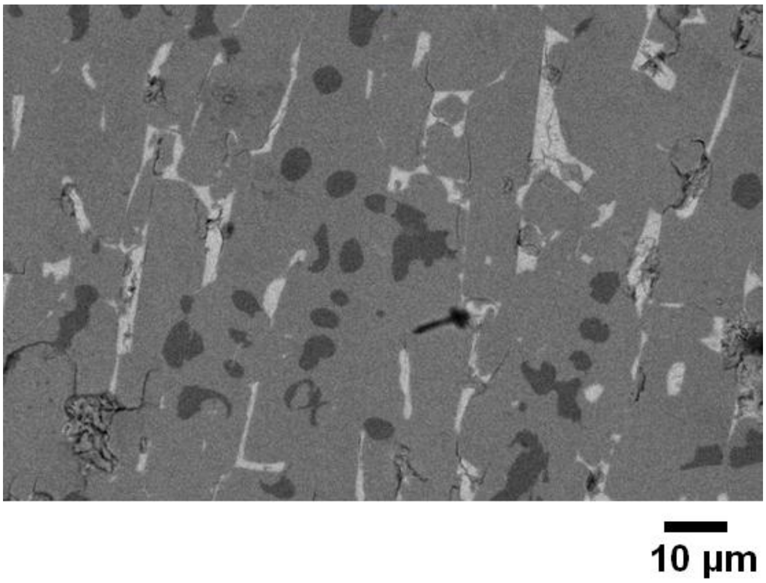

Figure 2 SEM-BSE image of Nd-Fe-B as-casted alloy

Low magnification SEM-BSE images of the as-casted alloy and different water-quenched alloys are shown in Figures 3a-3d, respectively. The as-casted alloy shows the segregation of the primary phases consisting of α-Fe phases 5-15 μm in size, $Nd_2Fe_{14}B$ phases as the matrix and Nd rich phases 10-30 μm in size. The water-quenched alloys show α-Fe phases that gradually decrease according to increasing soaking temperatures prior to water quenching. At 1200^{o}C the water-quenched alloy shows α-Fe phases like spider webs. Also, it can be seen that the Nd rich grain boundary phases gradually change from the triple junction grain boundary phases into the grain boundary phases.

Figures 4a-4d shows the SEM-BSE images of as-casted powder and water quenching followed by HDDR processed powders, respectively. It can be seen that the HDDR process changed the microstructure into almost a completely single $Nd_2Fe_{14}B$ phase. The HDDR process consists of four steps: (1) hydrogenation of $Nd_2Fe_{14}B$; (2) disproportionation of $Nd_2Fe_{14}B$ into α-Fe, NdH_2, and Fe_2B; (3) desorption of hydrogen gas from NdH_2; and finally (4) recombination of α-Fe, Nd, and Fe_2B into the $Nd_2Fe_{14}B$ (Ahmed et al., 2008; Sepehri-Amin et al., 2010; Nakamura et al., 1995; Sugimoto et al., 1999). During this process grain refinement occurs, resulting in $Nd_2Fe_{14}B$ single domain micro grain phases. The final phases after the HDDR process were $Nd_2Fe_{14}B$ phases and a small amount of Nd rich phases, resulting in a Nd phase that was absent in the recombination process. This Nd rich phase has an enormous influence on the magnetic properties of $Nd_2Fe_{14}B$.

The HDDR process is a method for producing a single domain $Nd_2Fe_{14}B$ phase within magnetic powders (Uehara et al., 1996). In the present HDDR processed samples it can be observed that the average grain size within the magnetic powder is about 0.3 μm, as can be seen in Figure 5. These grain sizes are in accordance with the theoretical single domain grain size.

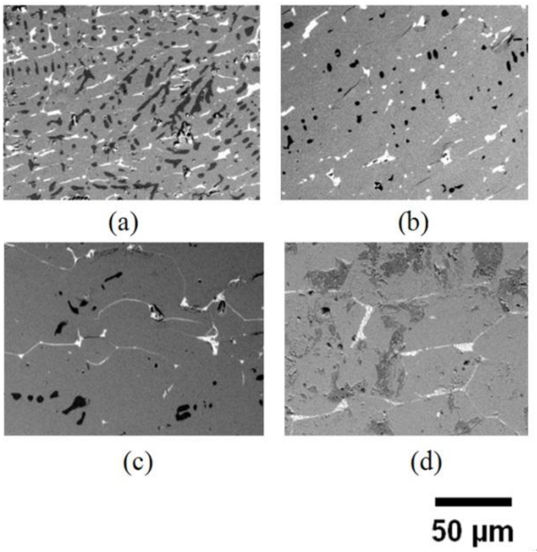

Figure 3 Low magnification of SEM-BSE images of: (a) the as-casted alloy and water-quenched alloys with different soaking temperatures; (b) 1000^{o}C; (c) 1100^{o}C; and (d) 1200^{o}C

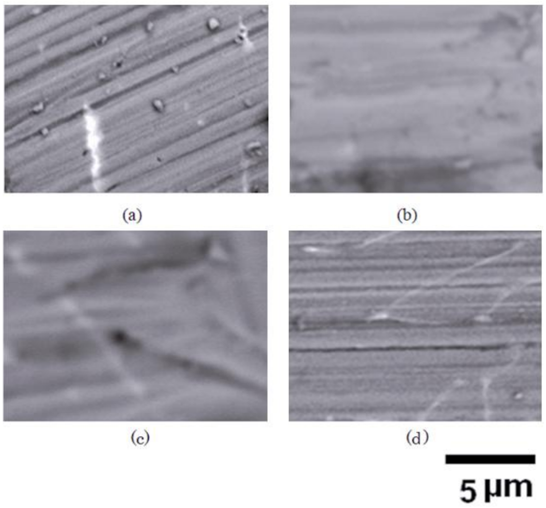

Figure 4 High magnification of SEM-BSE images of: (a) the as-casted alloy followed by HDDR processing, and water-quenched alloys with different soaking temperatures followed by HDDR processing; (b) 1000^{o}C; (c) 1100^{o}C; and (d) 1200^{o}C

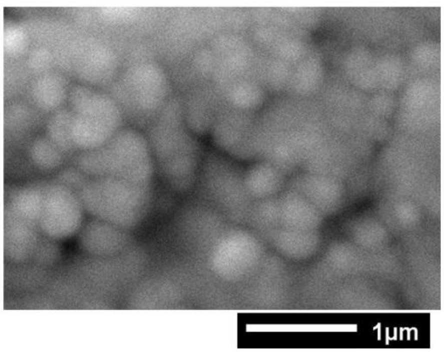

Figure 5 High magnification of SEM-BSE images of the HDDR-processed as-casted powder that shows the single domain of $Nd_2Fe_{14}B$ grains

4. DISCUSSION

The concentration of the Nd rich phase analyzed by the EDS intensity profiles was estimated to be around 98 wt. % (93 at. %). To explain the microstructural change, a metastable Fe-Nd diagram was used (Zhang et al., 1998). According to this diagram, the $Nd_{98\ wt.\%}$ rich phase starts to melt at 860°C. Increasing the temperature would decrease the cohesive force between the Nd atoms in the molten stage. The microstructure of HDDR processed as-casted powder shows a $Nd_2Fe_{14}B$ phase with some Nd rich triple junction micro grains from 1–2.5 µm. But the microstructure of 1000°C water-quenched powder is virtually free of the Nd rich phase. It is predicted that the Nd melted at 1000°C and subsequently diffused into the micro grain boundary phase, and therefore was not seen in the SEM-BSE image.

Increasing the temperature decreased the cohesive force so that the Nd rich phase diffused far away into the inter-grain boundary. In the 1100°C water-quenched powder most of the Nd rich phases diffused into the inter-micro grain boundary, and at 1200°C water-quenched powder the whole of the Nd rich phases diffused into the inter-micro grains. Figure 6 shows a schematic illustration of the Nd rich phase in the microstructure of magnetic powders.

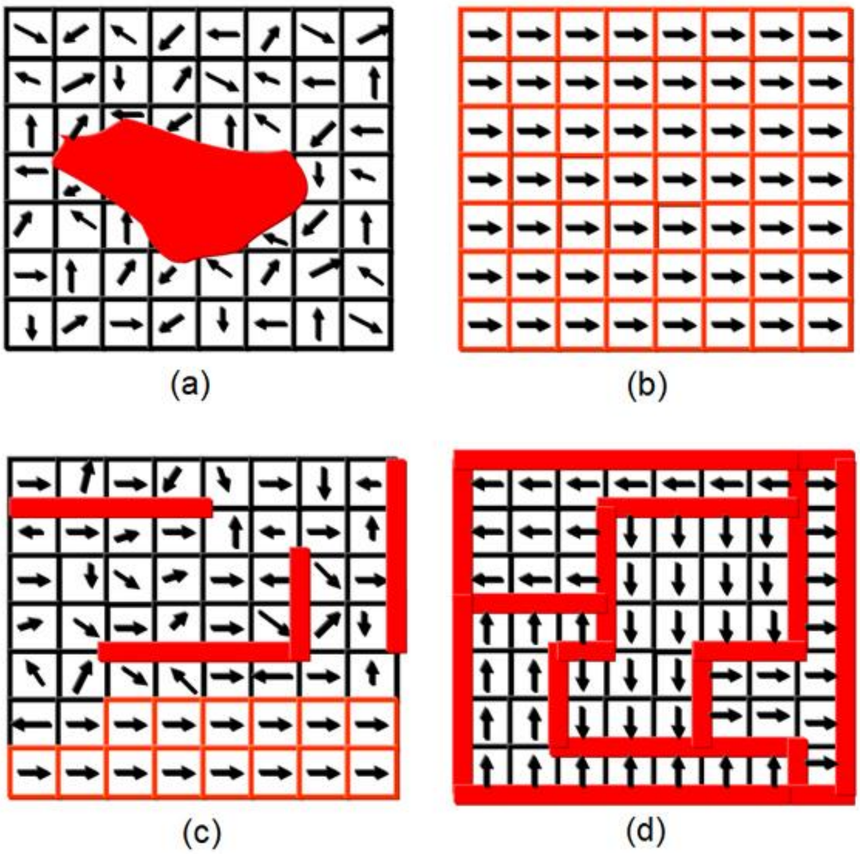

Figure 6 Schematic illustration of the Nd rich phase in the microstructure of: (a) the as-casted alloy followed by HDDR processing, and water-quenched alloys with different soaking temperature followed by HDDR processing; (b) 1000°C; (c) 1100°C; and (d) 1200°C

Figure 7 shows the typical magnetization curves, and Figure 8 shows the relationship between different processes and the magnetic coercivity of HDDR-processed samples. In the HDDR processed 1000°C water-quenched powder the coercivity increased rapidly, and subsequently decreased rapidly. It can be shown that the HDDR processed 1000°C water-quenched powder has the highest coercivity value.

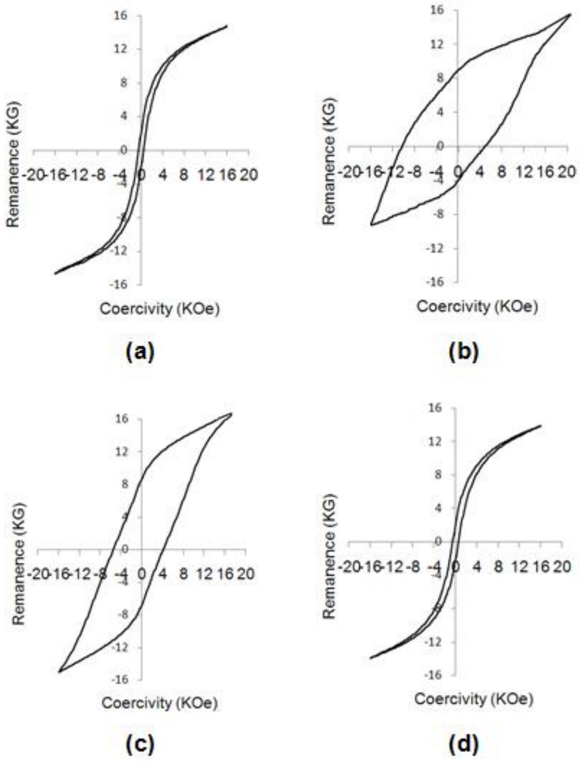

Figure 7 Magnetization curves for: (a) the as-casted alloy followed by HDDR processing, and water-quenched alloys with different soaking temperatures followed by HDDR processing; (b) 1000°C; (c) 1100°C; and (d) 1200°C

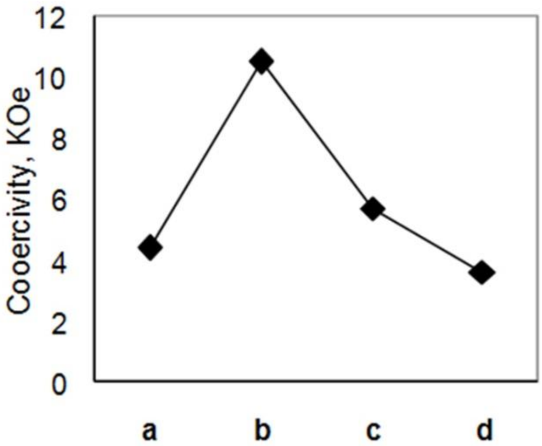

Figure 8 The relationship between different process conditions and coercivity: (a) The as-casted alloy followed by HDDR processing, and water-quenched alloys with different soaking temperatures followed by HDDR processing; (b) 1000°C; (c) 1100°C; and (d) 1200°C

In case the grain boundary phase contains a large amount of ferromagnetic elements, the $Nd_2Fe_{14}B$ grains would be exchange coupled, and the magnetic domain would extend to neighboring grains. But if the grain boundary contains a large amount of the Nd rich phase, the magnetic domain wall will be pinned with the Nd rich phase grain boundary. To increase the coercivity one should decrease the amount of ferromagnetic elements in the grain boundary phase, or in other words increase the amount of Nd rich phase to weaken the inter-grain exchange coupling (Sepehri-Amin et al., 2010; Sepehri-Amin et al., 2011).

As seen in Figure 4 and Figure 6, the Nd rich phase in the 1000^{o}C water-quenched magnetic powder diffused into the micro grain boundary single domain $Nd_2Fe_{14}B$ phase. The Nd rich phase that diffused into the micro grain boundary phase weakened the inter-micro grain exchange coupling, caused magnetic domain alignment, and finally increased the coercivity. Figure 9 compares the second quadrants of the magnetization curves of the 1000^{o}C water-quenched alloy and the homogenized alloy. The homogenized process was conducted for 12 h at 1100^{o}C in vacuum conditions It can be seen that the remanence and coercivity values for the water-quenched alloy and the homogenized alloy are 7.8 KG and 8.9 KG, respectively, and 12.6 KOe and 10.3 KOe, respectively. The water-quenched alloy has slightly higher remanence, but slightly lower coercivity. In general, both samples have similar magnetic properties.

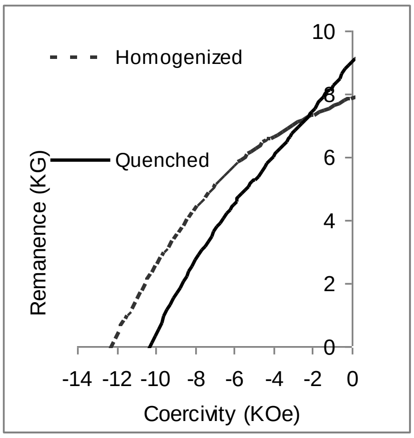

Figure 9 The second quadrant of magnetization curves of 1000^{o}C water-quenched and homogenized alloy magnetic powders, both followed by HDDR processing

5. CONCLUSION

The 1000^{o}C water quenching alloy followed by HDDR has similar magnetic properties compared to the homogenized alloy followed by HDDR. The microstructure of the former alloy was virtually a $Nd_2Fe_{14}B$ phase with a very slightly Nd rich phase that diffused into the micro grain boundary phase, increasing the coercivity.

6. ACKNOWLEDGEMENT

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