

Anisotropy of ternary and (Zr, Ga) addition magnetic powders

Min Liu*

Department of Electronic Science, Huizhou University, Huizhou 516007, Guangdong, China

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Abstract. Both non-homogenization treatment Zr, Ga addition and ternary Strip Casting alloy flakes can be used to prepare anisotropy HDDR NdFeB magnetic powders. This illustrates that anisotropy formation of magnetic powders neither depends on element addition nor depends on homogenization heat treatment of SC flakes. But the HDDR process procedure also plays an important role in anisotropy inducement. Highly anisotropic magnetic powders are attributed to rapid disproportionation reaction course, slow desorption-recombination reaction course, and optimum recombination hydrogen pressure during HDDR procedure process. This paper will provide an important guidance for preparing highly anisotropy magnetic powders with low cost.

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Key words: magnetic powders, SC alloy flakes, HDDR process, anisotropy

1 Introduction

The HDDR process (hydrogenation, disproportionation, desorption and recombination) has attracted broad attention for producing anisotropic Nd-Fe-B magnetic powders. Early experiment results indicated that addition of elements such as Co, Zr, Nb and Ga is prerequisite for anisotropy inducement in NdFeB-type alloys treated by the HDDR process [1]. The subsequent experiments showed that the anisotropic magnetic powders can also be obtained from purely ternary alloy subjected to an optimum HDDR process treatment [2-5]. Not only the elements addition, but also the HDDR process plays an important role in anisotropy formation. The HDDR magnetic powders are commonly prepared from segregated master ingots, and their magnetic properties are low due to the existence of soft magnetic phase α -Fe in master ingots. The SC (strip casting) alloy flakes have a good

*Corresponding author. *Email address:* liumin19811001@163.com (M. Liu).
Tel.: +86 7522527271; fax: +86 7522527271.

columnar crystalline structure, the main phase $Nd_2Fe_{14}B$ is uniformly separated by the symmetrical Nd-rich phase fine lamella and α -Fe is nonexistent, which is very suitable for preparing HDDR magnetic powders [6]. The HDDR magnetic powders prepared directly from non-heat-treated SC alloy flakes are isotropic [7]. But the anisotropic magnetic powders can be prepared from heat-treated SC alloy flakes subjected to the HDDR process [8]. Up to date, element addition and homogenization heat treatment of SC flakes are necessary condition or not for anisotropy formation, which has still not been reported. Based on current research situation, This paper tries to prepare anisotropic magnetic powder by using non-heat-treated Zr, Ga addition and ternary SC alloy flakes.

2 Experimental procedure

The $Nd_{12.8}Fe_{72}Co_{7.8}B_7Zr_{0.1}Ga_{0.3}$ and $Nd_{13.5}Fe_{79.5}B_7$ alloys are prepared using induction melting under argon atmosphere. The crushed alloys are put into the quartz tube of SC equipment waiting for melting, and then the molten alloys are ejected onto a copper wheel surface rotating at a speed of 3m/s to form the SC alloy flakes. The non-heat-treated SC alloy flakes are carried out a modified HDDR process to prepare the NdFeB magnetic powders. And the modified process schematic diagram is shown in Fig.1. First, the SC flakes are heated from room temperature to 800°C at a heating rate of 15°C/min in the hydrogen pressure of 1×10^5 Pa, and then kept at this temperature and pressure for 10 min-5 h. Second, the disproportionation products are followed by a slow desorption reaction at 850°C for 30min under hydrogen pressure of 10-70 kPa, and then followed by a fast desorption reaction in the high vacuum of 5×10^{-3} Pa at 850°C for 1h. Last, the alloys are quenched to room temperature in the argon atmosphere pressure of 1×10^5 Pa. After the magnetic powders are aligned in a magnetic field of 1.2 MA m^{-1} , their properties are measured using the vibrating sample magnetometer (VSM) with a maximum field of 2 MA m^{-1} at room temperature. The alignment degree of anisotropy (DOA) of the magnetic powders is evaluated using the ratio of remanence to saturation magnetization (B_r/B_s , the value of B_s is the actually measured value of the magnetic powders).

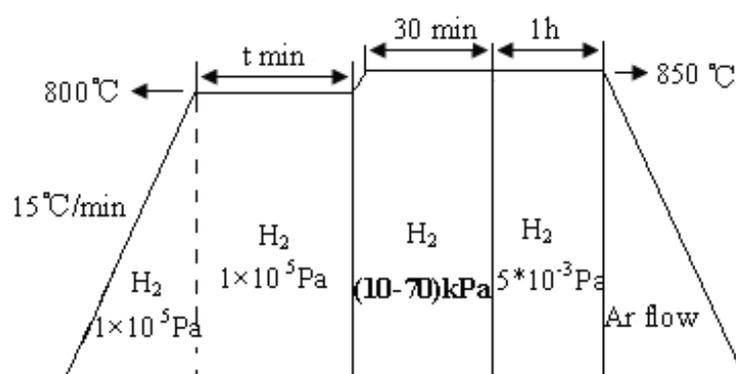


Figure 1: HDDR process schematic drawing.

3 Results and discussion

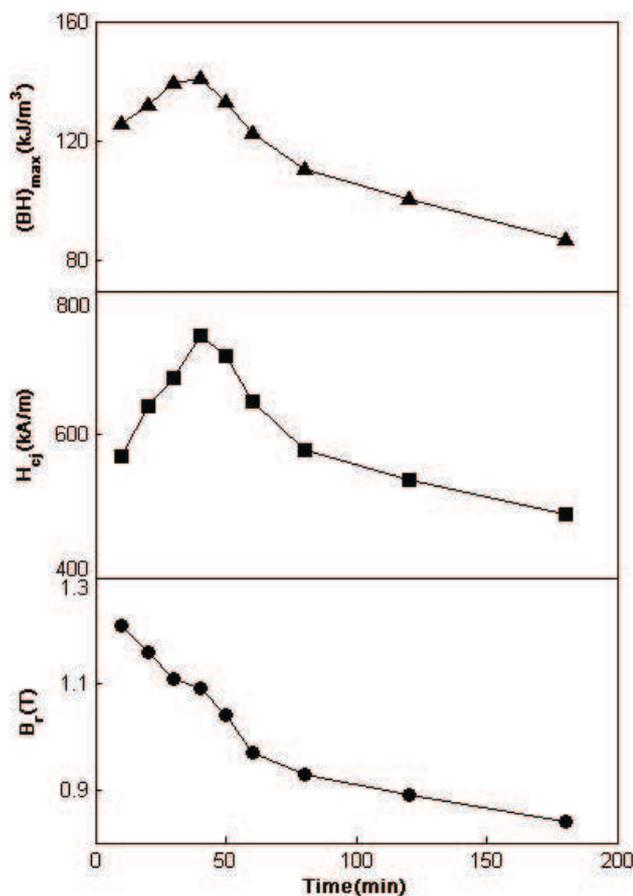


Figure 2: Dependence of magnetic properties on disproportionation time for Co, Zr, Ga addition powders.

Fig. 2 shows the effects of disproportionation time on the magnetic properties of Co, Zr, Ga addition powders, for disproportionation temperature of 800°C, and hydrogen pressure of 50 kPa during slow recombination stage. It can be seen that with increasing disproportionation time, both coercivity H_{cj} and magnetic energy product $(BH)_{max}$ increase firstly, and then decrease, but remanence B_r decreases. For disproportionation time of 40 min, the magnetic properties achieve maximum value of $B_r=1.09$ T, $H_{cj}=739.6$ kA/m, $(BH)_{max}=141.3$ kJ/m³. This is due to that for a short disproportionation time, the main phase and rich-Nd phase can't fully absorb hydrogen. But for a long disproportionation time, the disproportionation product dehydrogenize difficultly, thereby, the main phase grains recombine incompletely. So, whether a short or a long disproportionation times are unfavorable for magnetic properties [9]. The better disproportionation treatment time is 40min for $Nd_{12.8}Fe_{72}Co_{7.8}B_7Zr_{0.1}Ga_{0.3}$ alloy. Gutfleisch [10] pointed

out that the better disproportionation treatment time is 15min for $Nd_{16.2}Fe_{78.2}B_{5.6}$ alloy. Liao [11] pointed out that the better disproportionation treatment time is 2.5h for $Nd_{12.6}Fe_{69.3}Co_{11.6}B_{6.0}Al_{0.5}$ alloy. The better disproportionation time is attributed to different alloy composition and HDDR process treatment.

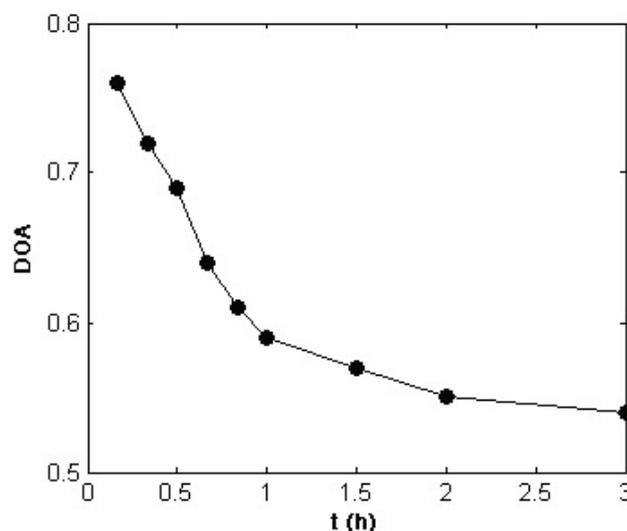


Figure 3: Effects of disproportionation time on alignment degree of anisotropy (DOA) for ternary powders.

Fig. 3 shows the dependence of alignment degree of anisotropy (DOA) on the disproportionation time, when the slow recombination hydrogen pressure is 20 kPa. For the disproportionation time of 10min, DOA is 0.76, the degree of crystallographic alignment is obvious. With increasing the disproportionation times, DOA gradually reduces. When the disproportionation time is close to 3 h, DOA is 0.54, the magnetic powders are almost isotropic. This illustrated that short disproportionation time is vital for the formation of anisotropy, but long disproportionation time is harmful for the anisotropy obtainment.

Respectively taking the optimum time of 40min for $Nd_{12.8}Fe_{72}Co_{7.8}B_7Zr_{0.1}Ga_{0.3}$ magnetic powder and of 10min for $Nd_{13.5}Fe_{79.5}B_7$ magnetic powder, Fig. 4 shows the dependence of respective degree of anisotropy DOA on the slow recombination hydrogen pressure ($HPSR$). It can be seen that with increasing $HPSR$, the DOA of both $Nd_{12.8}Fe_{72}Co_{7.8}B_7Zr_{0.1}Ga_{0.3}$ and $Nd_{13.5}Fe_{79.5}B_7$ magnetic powders increase firstly, and then decrease. While the $HPSR$ is 30 kPa, the DOA of both achieves the maximum values of 0.87 and 0.83, respectively. The ratio of remanence to saturation magnetization is much larger than 0.5, which indicates that These two magnetic powders are obviously anisotropic. It is assumed that the slow recombination rate (RR) only depends on the $HPSR$. For the $HPSR$ of 30 kPa, the RR is low, which is beneficial to the preferred orientations of anisotropy nucleus, and is also helpful to the improvement of magnetic properties in thermodynamic terms. Too low values of $HPSR$ quickens the RR . The faster RR is not only harmful for the formation of anisotropy. This is attributed to the assumption that

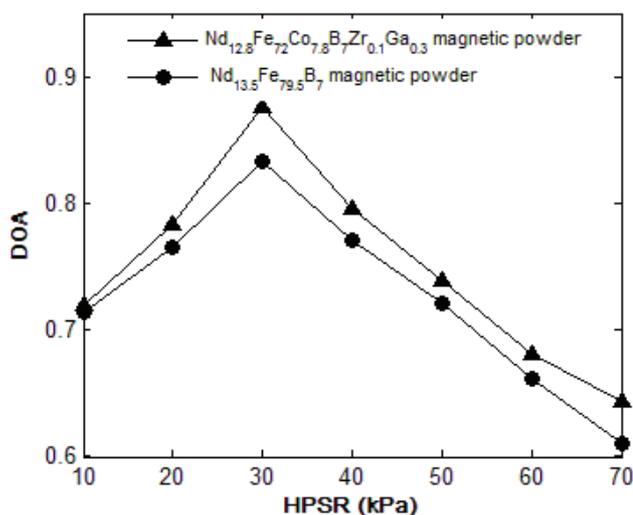


Figure 4: Dependence of degree of anisotropy DOA on slow recombination hydrogen pressure (HPSR) for $Nd_{12.8}Fe_{72}Co_{7.8}B_7Zr_{0.1}Ga_{0.3}$ and $Nd_{13.5}Fe_{79.5}B_7$ magnetic powders.

the faster RR will lead to random orientations of recombined grains [12]. Too high values of HPSR lowers the RR, and the lower RR will lead to the excessive growth of newly recombined $Nd_2Fe_{14}B$ grain, and appearance of large grains with irregular boundaries. Both short and long time will lead to the decrease of degree of anisotropy.

Figs. 5(a) and 5(b) show the microstructures of $Nd_{12.8}Fe_{72}Co_{7.8}B_7Zr_{0.1}Ga_{0.3}$ and $Nd_{13.5}Fe_{79.5}B_7$ magnetic powder, respectively, for HPSR of 30kPa. It can be seen that the grain size equally distributes for both magnetic powders, which approximately equals

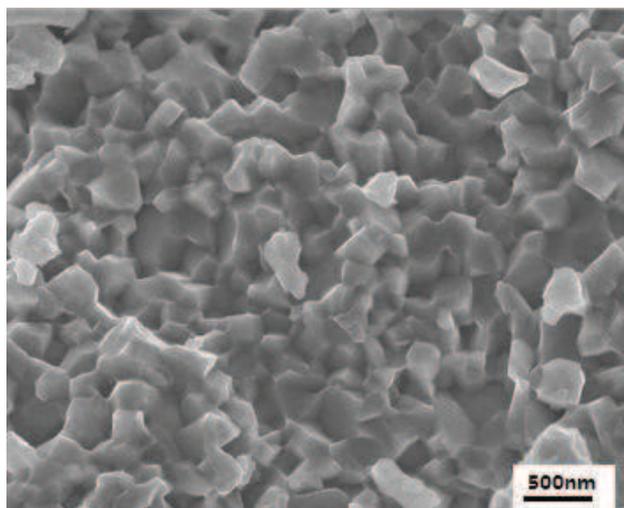


Figure 5: (a) Microstructures of $Nd_{12.8}Fe_{72}Co_{7.8}B_7Zr_{0.1}Ga_{0.3}$ magnetic powder.

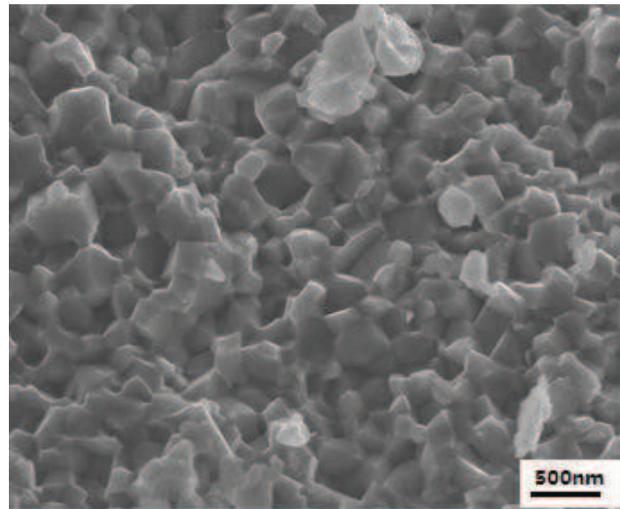


Figure 5: (b) Microstructures of $Nd_{13.5}Fe_{79.5}B_7$ magnetic powder.

to 300nm. At the same time, the properties of these two magnetic powders achieve maximum values, respectively. the properties of $Nd_{12.8}Fe_{72}Co_{7.8}B_7Zr_{0.1}Ga_{0.3}$ magnetic powder achieve values of $B_r=1.3$ T, $H_{c_j}=954.3$ kA/m, $(BH)_{max}=259$ kJ/m³, and the properties of $Nd_{13.5}Fe_{79.5}B_7$ magnetic powder achieve values of $B_r=1.24$ T, $H_{c_j}=875.4$ kA/m, $(BH)_{max}=216$ kJ/m³.

The anisotropy purely ternary and Co, Zr, Ga addition magnetic powders are prepared by using the same process route, but adopting different process parameters. For different ingredient, the experiment conclusion is the same. The investigation indicates that whether for the purely ternary or to Co, Zr, Ga addition permanent materials, as long as the disproportionation time is optimum, the effect of slow recombination hydrogenation pressure on the anisotropy and magnetic properties is very obvious. The highly anisotropic magnetic powders can be obtained if the hydrogen pressure can be controlled in the appropriate range. Simultaneously, the investigation also confirms that the anisotropic magnetic powders can be prepared directly from purely ternary and from Zr, Ga addition non-heat-treated SC alloy flakes (The magnetic properties of powders from Zr, Ga addition alloy flakes are higher than that of from the ternary alloy flakes treated by the HDDR process). The above conclusion indicates that the element added and SC flakes subjected to the homogenization heat treatment are not prerequisite for obtaining highly anisotropic magnetic powders. But the HDDR process adjustment is the key factor for the anisotropy formation. So long as to speed disproportionation reaction course, slow desorption-recombination reaction course, and to control recombination hydrogen pressure, the highly anisotropic magnetic powders can be prepared.

4 Conclusions

Anisotropy NdFeB magnetic powders can be obtained both from non-homogenization treatment Zr, Ga element addition and purely ternary SC alloy flakes. The SC+HDDR process procedure is simple, and is easy to operate, therefore, it provides an important guidance for preparing highly anisotropy NdFeB magnetic powders with low cost for manufacturing enterprises, especially for magnetic powder manufacturing enterprise.

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