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Research articles

# Effect of hydrogen pressure on hydrogen absorption of waste Nd-Fe-B sintered magnets

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ABSTRACT

Hydrogen treatment technology is developed to recycle the waste Nd-Fe-B sintered magnets in the way of short route, low cost, and high efficiency. In present study, effect of hydrogen pressure on hydrogen decrepitation (HD) process of sintered Nd-Fe-B strip casting flakes (SC) and waste sintered magnets (SM) was systematically studied. Both SC and SM show the approximate HD process, which accelerates with increasing pressure. The surface activation process accelerates with increasing pressure, and almost disappears when the initial hydrogen pressure reaches to 6 MPa. The hydrogen content of SM is lower than SC, and increases with increasing pressure. For the SM, blasting power of HD process decreases with the increase of the initial hydrogen pressure. It provides beneficial reference for recycling waste Nd-Fe-B sintered magnets.

## 1. Introduction

In recent years, Recycling of waste Nd-Fe-B sintered magnets has drawn tremendous attention due to the rapid growth of demand for Nd-Fe-B magnets and volatility in the price of rare earth elements such as Nd and Dy [1–5]. It should be noted that hydrogen plays a vital role in recycling of waste sintered Nd-Fe-B magnets. Sheridan et al. investigated sintered Nd-Fe-B-based scrap magnets were recovered and processed using hydrogen decrepitation (HD) and hydrogenation-disproportionation-desorption-recombination (HDDR) routes [6]. Zakotnik et al. produced recycled sintered magnets from anisotropic powders recovered from Nd-Fe-B sintered magnets by HD process [7]. Luo et al. reported anisotropic powders were prepared by hydrogen absorption and desorption of high coercivity NdDyFeCoNbCuB sintered magnets [8]. In our previous study, the waste Nd-Fe-B sintered magnets were recycled to manufacture sintered magnets by combination of hydrogen decrepitation and grain boundary modification techniques [5].

Nowadays, HD process is an important procedure in the manufacture of Nd-Fe-B sintered magnets [9–11]. The Nd-Fe-B alloy absorbs hydrogen very readily at room temperature with the consequent decrepitation of the bulk material into a friable powder. The absorption process is consist of two stages whereby hydrogen is first absorbed by the Nd-rich grain boundary phase and then by the matrix $Nd_2Fe_{14}B$ phase. Eqs. (1) and (2) corresponds to the absorption of hydrogen by the Nd-rich phase and $Nd_2Fe_{14}B$ phase, respectively, where $x$ and $y$ are dependent on temperature and pressure [6,12].

$$Nd + \frac{x}{2}H_2 = NdH_x \tag{1}$$

$$Nd_2Fe_{14}B + \frac{y}{2}H_2 = Nd_2Fe_{14}BH_y \tag{2}$$

Up to now, HD has been well developed as an effective technique to fabricate Nd-Fe-B type powders using textured ingots or strip casting flakes because HD powders can be prepared without damaging the textured microstructure by using this technique [13,14]. However, reports on hydrogen absorption of waste Nd-Fe-B sintered magnets are rare. In present study, effect of hydrogen pressure on HD process for Nd-Fe-B strip casting flakes (SC) and waste sintered magnets (SM) were systematically studied. Moreover, the hydrogen content and blasting power of HD process were investigated.

## 2. Experimental

The Nd-Fe-B SC and SM employed in the present studies are commercial grade 42H. The element contents of specimens were established by inductively coupled plasma (ICP) analysis (Prodigy XP). Table 1 shows the element contents of the SC and SM. Rare earth (RE) contents of SM are less than SC, it is because RE elements evaporate in the

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**Table 1**
Element contents of the SC and SM (wt.%).

| Element | SC | SM |
|---|---|---|
| Fe | 66.23 | 67.13 |
| Nd | 23.23 | 23.02 |
| Pr | 7.36 | 6.63 |
| Co | 0.79 | 0.87 |
| Ho | 0.55 | 0.52 |
| Al | 0.27 | 0.24 |
| Zr | 0.25 | 0.24 |
| Cu | 0.17 | 0.2 |
| Ga | 0.17 | 0.17 |
| B | 0.98 | 0.98 |
| ΣRE | 31.14 | 30.17 |

sintering process for magnets from SC.

The thickness of SC is about 0.3 mm and SM is crushed into small scraps (D < 10 mm per piece). The HD process for the specimens of 200 g were tested in an high temperature and high pressure autoclave (Fike P-ST-FS) at 288 K with initial hydrogen pressure of 0.1–15 Mpa. The total amount of hydrogen in the autoclave is low under the initial hydrogen pressure of 0.1 Mpa, the autoclave need refill with hydrogen. The hydrogen contents of the specimens were measured by pulse heating inert gas method of infrared absorption (QB-QT-38-2014). Cell volume and lattice parameters of Nd-Fe-B HD powders were investigated by X-ray diffraction (XRD).

## 3. Results and discussion

### 3.1. HD process

Fig. 1 shows the hydrogen decrepitation curves of Nd-Fe-B SC and SM at 288 K with different initial hydrogen pressure. Under the initial hydrogen pressure of 0.1–1 Mpa, the HD process composes of four stages of magnets surface activation, slow hydrogenation of Nd-rich grain boundary phase, quick hydrogenation of $Nd_2Fe_{14}B$ main phase grains, and slow hydrogenation of inner part of the magnets. The surface activation stage of SM is shorter than SC, because the surface of SM scraps is rougher than SC and the surface defects accelerate physical adsorption for hydrogen. The Nd-rich phase expands and cracks in the hydrogenation of Nd-rich grain boundary phase. The morphology of SC and SM is shown in Fig. 2. The cracks of SM are more than SC because the Nd-rich phase of SM is thicker than SC. Therefore, the hydrogenation of Nd-rich grain boundary phase of SM is faster than SC. The hydrogenation of $Nd_2Fe_{14}B$ alloy is an exothermic reaction resulting in blasting. The hydrogenation of $Nd_2Fe_{14}B$ main phase grains of SM is faster than SC because the blasting power of HD process for SM is stronger than SC. The inner part of SM is larger than SC, so hydrogenation of inner part of SM is slower than SC.

The surface activation and HD process accelerate with increasing hydrogen pressure, because physical adsorption for hydrogen and hydrogenation of specimens accelerate with increasing hydrogen. Under the initial hydrogen pressure of 6–15 Mpa, the surface activation almost disappears, the stages of hydrogenation of Nd-rich grain boundary phase and $Nd_2Fe_{14}B$ main phase grains combine. When the initial hydrogen pressure is higher than 6 MPa, the hydrogen pressure drops rapidly at the beginning, indicating that the hydrogenation of Nd-rich phase and $Nd_2Fe_{14}B$ phase rapidly takes place. In the stages of hydrogenation of inner part, the pressure drops slowly and the reaction rate decreases.

### 3.2. The hydrogen content

Fig. 3 shows the hydrogen content of SC and SM at 288 K with initial hydrogen pressure of 0.1–15 Mpa. Under the initial hydrogen pressure of 0.1–0.5 Mpa, the hydrogen content of SC and SM is 0.43 wt% and 0.38 wt%, respectively, indicating that it's not dependent on pressure. When the initial hydrogen pressure is higher than 1 MPa, the hydrogen content slightly increases. The hydrogen content of SC and SM is 0.432 wt% and 0.404 wt%, respectively, when the initial hydrogen pressure is 15 Mpa. The hydrogen content of SM is lower than SC, it is because rare earth contents of SM are lower than SC, Table 1 shows the rare earth contents of SC and SM are 31.14 wt% and 30.17 wt%, respectively.

Fig. 4 is XRD patterns of SC and SM HD powders. The peaks of $Nd_2Fe_{14}BH_x$ is the same as $Nd_2Fe_{14}B$, however, the peak position moves

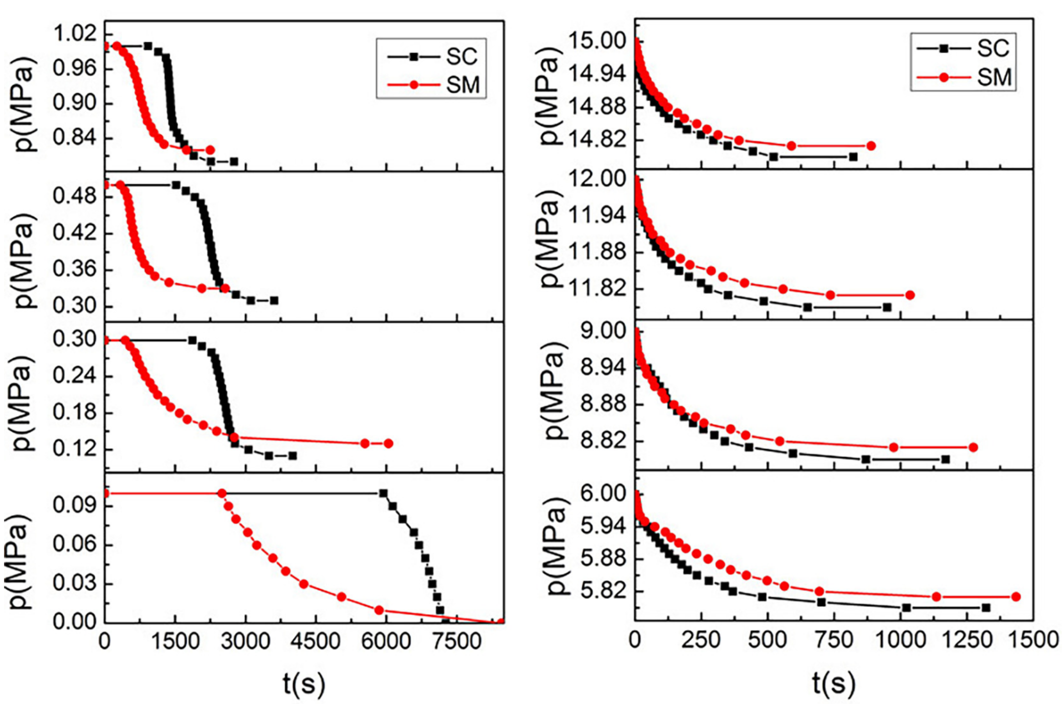


**Fig. 1.** Hydrogen decrepitation curves of SC and SM at 288 K with initial hydrogen pressure of 0.1–15 Mpa.