

Pressure-temperature Diagram of Critical Condition for Hydrogen-assisted Disproportionation of Nd-Fe-B Alloy

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1. Introduction

A Nd-Fe-B permanent magnetic alloy exhibits virtually no coercivity in cast ingot condition primarily because of the coarse grain size of the Nd₂Fe₁₄B hard magnetic phase in the alloy. The most effective way of converting the no coercivity ingot material to a highly coercive one is the HDDR (hydrogenation, disproportionation, desorption and recombination) process. Through the HDDR treatment, the coarse Nd₂Fe₁₄B grains in the alloy ingot is re-structured to fine grains, of which size is comparable to the critical single domain size ($d_c \approx 0.3$ μm) of the Nd₂Fe₁₄B phase. More interestingly, a carefully controlled HDDR process can lead to an anisotropic powder with good Nd₂Fe₁₄B grain texture. In the anisotropic powder, the newly formed fine grains are oriented orderly (texture) in such a manner that the magnetisation easy axis (c-axis) of the fine Nd₂Fe₁₄B grains are parallel to each other maintaining the crystallographic orientation of their mother grain. In this HDDR treatment leading to an anisotropic powder, the critical step for inducing the texture is the disproportionation, and of which kinetics is controlled to be as slow as possible. This is commonly accomplished by employing a lower hydrogen pressure. In the present study, the critical condition (hydrogen pressure and temperature) for the disproportionation reaction of a hydrided Nd-Fe-B alloy was investigated.

2. Experimental Work

The Nd_{12.5}Fe_{81.1-(x+y)}B_{6.4}Ga_xNb_y (x=0 or 0.3, y = 0 or 0.2) alloys used in the present study were prepared by induction-melting of the high purity constituent metals. The prepared alloy ingots were homogenised at 1413 K for 40hr under argon gas, and pulverised into powder with a particle size of 50-150 μm . The powder material (350 mg) was placed into thermopiezic analyser (TPA) chamber with approximate volume of 160 cm^3 , and the chamber was evacuated and then filled with hydrogen ($p = 0.1$ MPa). The charged material was first fully hydrogenated in the TPA by heating it in hydrogen gas up to 400°C (heating rate = 7°C/min) and holding at there for 1 hr. Immediately after the completion of full hydrogenation, the hydrogen pressure in the chamber was swiftly adjusted to a desired pressure, and the hydrogenated material was further heated toward 820°C from 400°C. In the course of heating from 400°C, the hydrogen pressure change with temperature was carefully monitored. Based upon these TPA results, a precise condition of hydrogen pressure and temperature for the disproportionation reaction was decided. In addition to the hydrogen gas atmosphere for the disproportionation, a mixed gas atmosphere of hydrogen and argon was also used. Magnetic phase analysis of the material at various conditions was performed by magnetic balance-type thermo-magnetic analyser (TMA).

3. Results and discussion

Hydrogen pressure – temperature diagram showing the critical condition for the disproportionation of the fully

hydrogenated $\text{Nd}_{12.5}\text{Fe}_{81.1-(x+y)}\text{B}_{6.4}\text{Ga}_x\text{Nb}_y$ ($x = 0$ or 0.3 , $y = 0$ or 0.2) alloys in different atmospheres of pure hydrogen and mixed gas of hydrogen and argon was established as shown in Fig. 1. In order for the fully hydrogenated $\text{Nd}_{12.5}\text{Fe}_{81.1-(x+y)}\text{B}_{6.4}\text{Ga}_x\text{Nb}_y$ ($x = 0$ or 0.3 , $y = 0$ or 0.2) alloys to be disproportionated, the reaction condition needs to be placed within the right-hand side of the curves. The onset temperature of disproportionation reaction of the fully hydrogenated $\text{Nd}_{12.5}\text{Fe}_{81.1-(x+y)}\text{B}_{6.4}\text{Ga}_x\text{Nb}_y$ alloys was slightly increased as the hydrogen pressure decreased. The addition of Ga and Nb led to a shift of critical disproportionation condition towards higher hydrogen pressure and higher temperature. The critical disproportionation condition of the hydrogenated Nd-Fe-B alloys shifted towards harsher condition in the mixed gas (hydrogen and argon) atmosphere than in the pure hydrogen gas. The hydrided materials were completely disproportionated after the heating up to 820°C regardless of alloy- and atmosphere-type when the hydrogen (or partial) pressure on the starting of disproportionation was higher than 300 mbar. However, when the hydrogen (or partial) pressure on the starting of disproportionation was lower than 200 mbar, the hydrided materials were only partially disproportionated after the heating up to 820°C regardless of alloy- and atmosphere-type.

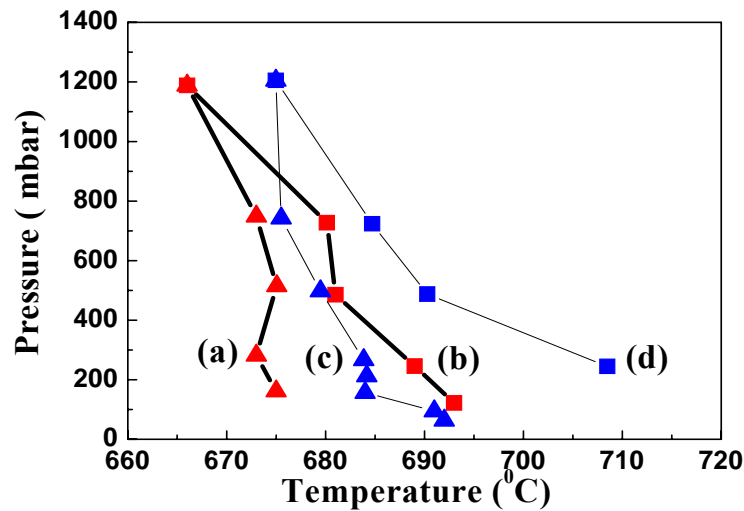


Fig. 1. Hydrogen pressure - temperature diagram showing the critical condition for the disproportionation of the fully hydrogenated $\text{Nd}_{12.5}\text{Fe}_{81.1-(x+y)}\text{B}_{6.4}\text{Ga}_x\text{Nb}_y$ alloys in different atmospheres. (a) $\text{Nd}_{12.5}\text{Fe}_{81.1}\text{B}_{6.4}$, hydrogen, (b) $\text{Nd}_{12.5}\text{Fe}_{81.1}\text{B}_{6.4}$, hydrogen + argon, (c) $\text{Nd}_{12.5}\text{Fe}_{80.6}\text{B}_{6.4}\text{Ga}_{0.3}\text{Nb}_{0.2}$, hydrogen, (d) $\text{Nd}_{12.5}\text{Fe}_{80.6}\text{B}_{6.4}\text{Ga}_{0.3}\text{Nb}_{0.2}$, hydrogen + argon.

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