



## RESEARCH ARTICLE

### THE EFFECT OF ADDING HEAVY RARE EARTH HYDRIDE ON MAGNETIC PROPERTIES OF NDFEB –TYPE SINTERED MAGNETS USING APPROPRIATE HEAT TREATMENT

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#### ABSTRACT

NdFeB type sintered magnets were subjected to the process of grain boundary diffusion and their magnetic properties and microstructures were analysed. The magnets were coated with a suspension of (Dy/Tb)H<sub>x</sub> and cyclohexane, which were then subjected to heat treatment. The coercivity was enhanced with a little reduction in remanence. The (Dy/Tb)H<sub>x</sub> was shown to have diffused into the magnet through the grain boundaries and formed a Dy/Tb rich shells around the Nd<sub>2</sub>Fe<sub>14</sub>B (matrix) phase. An increase of just over 300kA/m of coercivity was achieved.

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## INTRODUCTION

Sintered magnets based on NdFeB, yield the highest value of (BH) *max* of any permanent magnet material. This has led to them being vital components in a widespread range of applications such as high-performance motors, actuators, generators and sensors. These materials utilise the strong magnetic moment arising from the Nd<sub>2</sub>Fe<sub>14</sub>B (Ø) phase. This phase also exhibits strong magneto-crystalline anisotropy, leading to high intrinsic coercivities. Control of the microstructure is vital in obtaining the desired magnetic properties (Endoh, 1994; Kaneko, 1994). Since the introduction of NdFeB magnets made by powder processing (Sagawa, 1984), a range of elements have been added to the basic composition in order to modify the properties. Most reported techniques have involved melting all the elemental components together to form an alloy of the desired magnet composition. Since the mechanism on magnetic domain reversal in NdFeB sintered magnets is strongly dependent on the nucleation of reverse domains, the microstructure in the vicinity of the grain boundary is very important in achieving this high coercivity by pinning magnetic domains. It is well known that the addition of heavy rare earths (Dy, Tb) is very effective in enhancing the coercivity because these heavy rare-earth element increases the magnetic anisotropy field of the (Nd, Dy/Tb)<sub>2</sub>Fe<sub>14</sub>B compound (Hirose et al., 1986; Koon, 1985). Some authors (Hong, 1995; Mottram, 1999;

Mottram, 2001; Zakotnik, 2008; Zakotnik, 2008) have used a range of powder blending techniques to modify the grain boundary composition by blending in alloys richer in Nd or modifying elements. These additions can then be used to modify the grain boundary or the surface of grains by diffusion process in which heavy rare earth oxide or fluoride powders can be used for coating the magnet (12-15). When comparing the two processes, it is possible to find advantages and disadvantages. For example, blending gives a more homogenous distribution of the heavy rare earth element throughout the grains of the matrix phase, and hence giving rise to an expected increase in coercivity. The diffusion process results in a minimum loss of remanence, but might be limited by the thickness of the processed magnet. The geological abundance of the heavy rare earths is very low compared to neodymium, and so consumption may become an issue in the future. The optimum NdFeB-type sintered magnets should therefore be made without or with minimal additions of the heavy rare earths elements and this represents the next challenging step for the magnet industry. In the present work, modified sintered magnets were prepared by a combination of heat treatments and the addition of various heavy rare earth elemental hydrides on the surface of sintered magnets. This opens an alternative route to modify the composition after sintering which can give a significant degree of freedom throughout the manufacturing process to tailor the magnetic properties. This process requires the addition of much less heavy rare earth elements and thus

helps to reduce the depletion of the natural resources, and is therefore essential to the environment and to production of sintered magnets.

## EXPERIMENTAL METHOD

The starting materials employed in the present work are in the form of sintered segments obtained from the VCM of hard disc drives. Each segment weighted  $\sim 10\text{g}$  and the nickel coating was removed by grinding off the coating by hand. Rectangular sections of the NdFeB magnets were then prepared with dimensions of  $14\text{ mm} \times 15\text{ mm} \times 2\text{ mm}$ . The  $\text{TbH}_x$  or  $\text{DyH}_x$  powder has been mixed with cyclohexane to form a suspension. This mixture was in the ratio of 2:1 in weight, with a mean particle size of  $3\text{--}5\text{ }\mu\text{m}$ . The sintered magnets were coated and dried in a glove box, housed in a stainless steel envelope and placed in a furnace. The magnets were heated at  $800\text{ }^\circ\text{C}$  for 0.5hr, 1hr, 3hrs and 10 hours under vacuum conditions of  $6 \times 10^{-2}\text{ Pa}$ . The magnetic properties were measured using a permeameter after pulse magnetisation with a  $4.5\text{T}$  field. The microstructure analyses of these magnets were performed using a SEM JEOL 7000.

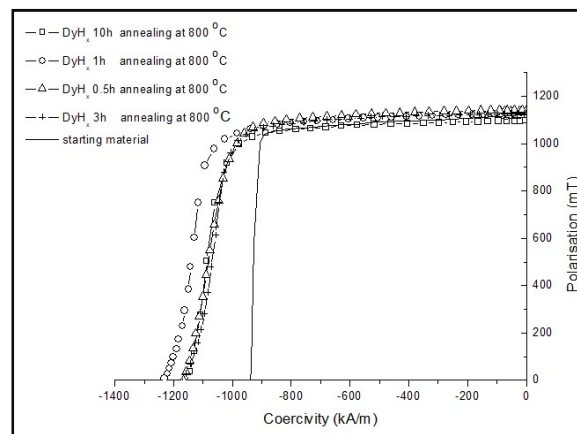
## RESULTS AND DISCUSSION

The initial sintered VCM magnets were analysed for composition by ICP at Less Common Metals. A typical composition is given in Table 1. It can be seen that the sintered magnets were quite rich in the rare earth component of Nd with a minor addition of Dy that improves coercivity component. There is also small addition of Al which improves the wettability of the grain boundary phase. A typical second quadrant demagnetisation curves is shown in Figure 1. The coercivity of the 0.5h, 1h, 3h and 10h, heat treated sintered magnets with  $\text{DyH}_x$  resulted in an increase by about 232, 391, 229, 232 kA/m, respectively. As expected, the addition of Dy resulted in a significant increase in the intrinsic coercivity, probably due to magneto-crystalline anisotropy. The remanence after the heat treatment process was on average reduced by 50 mT. When the compared  $\text{DyH}_x$  heat treatment process is compared with the blending of the  $\text{DyH}_x$  to the sintered magnets, the latter results in a dramatic decrease of remanence. The loss of remanence is ascribed to the antiparallel coupling with the Fe of the Dy in the  $\phi$ -phase ( $\text{Nd}_2\text{Fe}_{14}\text{B}$ ), which has a dramatic impact when  $\text{DyH}_x$  is blended into the virgin material which is due to the large magnetic moment (Dy) and to the more homogeneous distribution of the Dy within the  $\phi$ -phase. These results are in agreement with previous work on DyH blending additions to sintered magnets; see for example (8, 9, and 11). Figure 2 shows a typical second quadrant demagnetisation curves for the  $\text{TbH}_x$  heat treated sintered magnet. The coercivity of the 0.5 h, 1 h, 3 h and 10 h, heat treated sintered magnets which resulted in an increase by about 267, 370, 406, kA/m, respectively and the 10 h anneal could not be measured due to insufficient demagnetising field. The coercivities of the heat treated sintered magnets with  $\text{TbH}_x$  are generally higher than these of heat treated sintered magnets with  $\text{DyH}_x$  because of the magnetic anisotropy field of  $(\text{Nd}, \text{Tb})_2\text{Fe}_{14}\text{B}$ , which is higher than that of  $(\text{Nd}, \text{Dy})_2\text{Fe}_{14}\text{B}$  (4). The summary of the magnetic properties for Dy and Tb heat treated are summarised in Figure 3. Generally it can be seen that, with the increasing values in coercivity, there is a marked fall in the remanence. It can be seen that one hour heat treated sintered magnets with

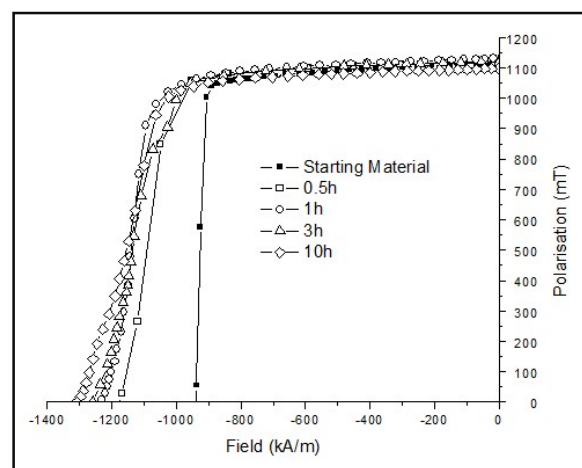
$\text{DyH}_x$  resulted in optimum magnetic properties. The decreased remanence value was insignificant while the coercivity was increased for a 390 kA/m.

**Table 1. ICP analysis (there are other minor additions not being included) of the typical composition of the sintered magnet ( $\pm 0.008\text{--}0.073$ )**

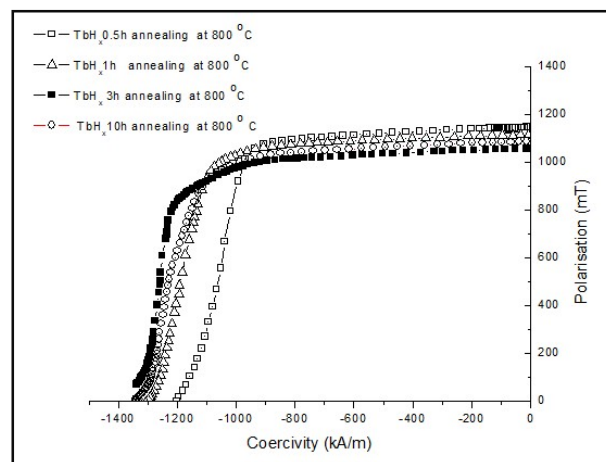
element	Nd	Dy	Fe	B	Al
at. %	13.78	0.63	77.81	6.44	0.69



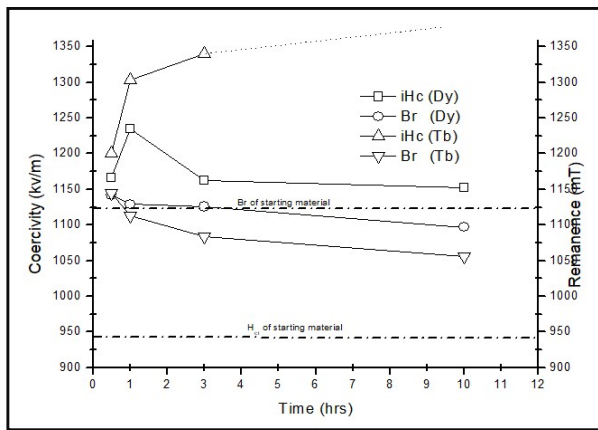
**Figure 1. Demagnetising curves for  $\text{DyH}_3$  heat treated magnets at  $800^\circ\text{C}$ , with various times**



**New graph of DyH**



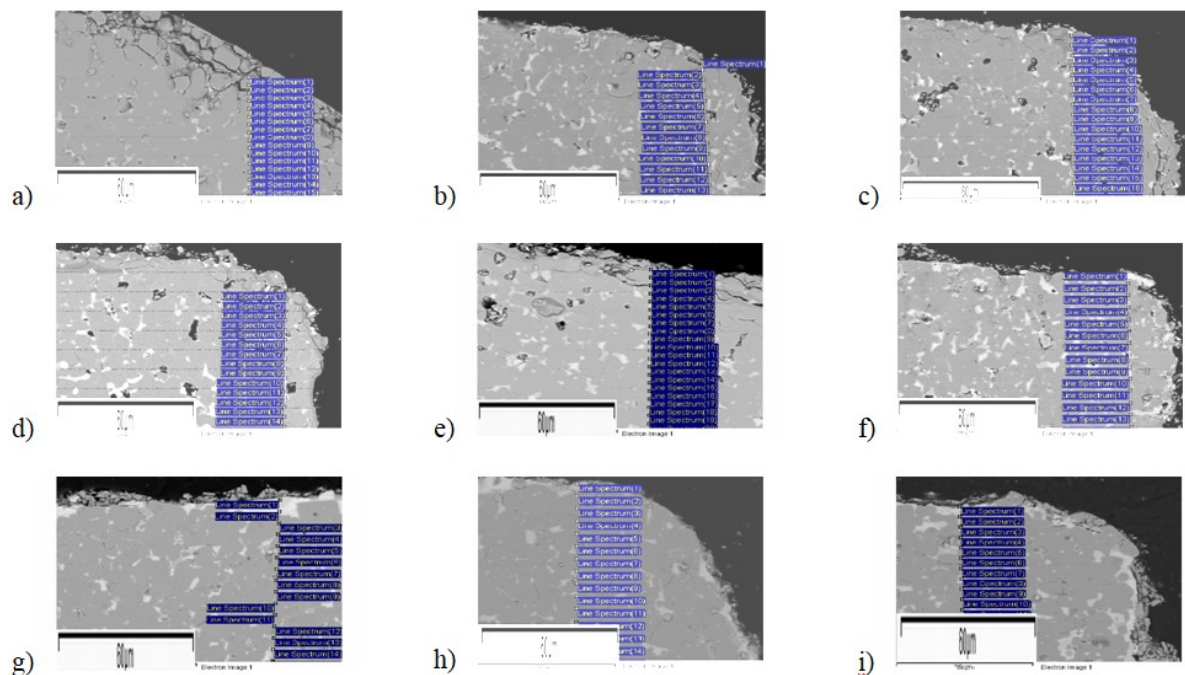
**Figure 2. Demagnetising curves for  $\text{TbH}_3$  heat treated magnets at  $800^\circ\text{C}$ , with various times**



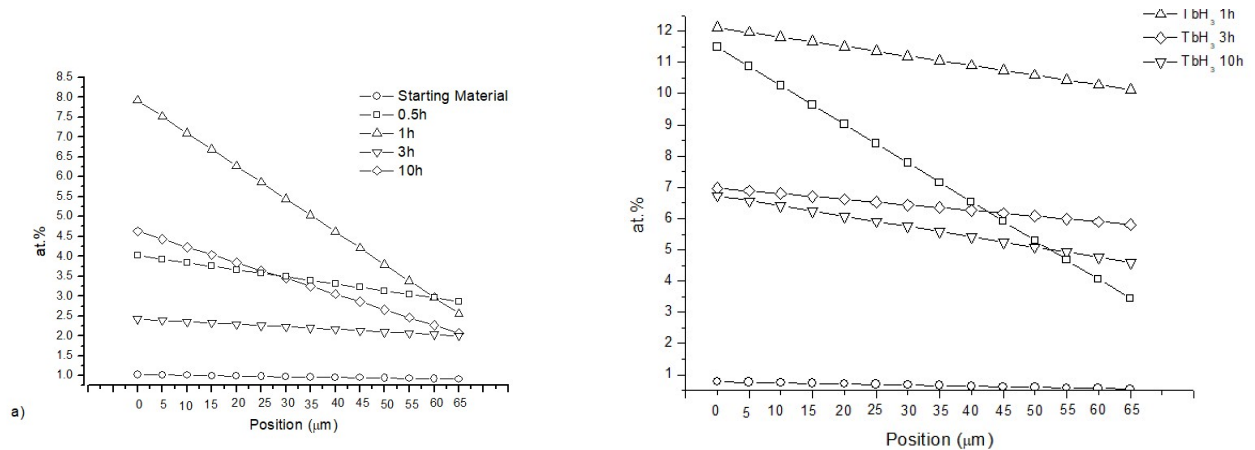
**Figure 3. Summary of magnetic properties of heat treated sintered magnets with Dy/Tb hydrides versus heat treatment times**

Table 1 ICP analysis (there are other minor additions not being included) of the typical composition of the sintered magnet ( $\pm 0.008-0.073$ ). Figure 4 summaries backscatter images of Dy/Tb hydrides. EDX analysis was performed from the edge of the heat treated magnets for a distance of  $60\ \mu\text{m}$ . Compositional profiles of Dy/Tb hydrides were taken from heat treated sintered magnets and these results are presented in Figure 5 (a) and (b).

From Figure 5a, it is apparent that the heat treated magnets with  $\text{DyH}_x$  show similar values of Dy content for a distance of  $60\ \mu\text{m}$ . When compared with figure 5b, the heat treated magnets processed with  $\text{TbH}_x$  from the surface almost doubled the amount of heavy rare earth element. The possible explanation for such behaviour might be that  $\text{TbH}_x$  is more reactive than  $\text{DyH}_x$ .



**Figure 4. Microstructures of heat treated sintered magnets at  $800^\circ\text{C}$ : (a) starting material, (b)  $\text{DyH}_3$  0.5 h, (c)  $\text{DyH}_3$  1 h, (d)  $\text{DyH}_3$  3 h, (e)  $\text{DyH}_3$  10 h, (f)  $\text{TbH}_3$  0.5 h, (g)  $\text{TbH}_3$  1 h, (h)  $\text{TbH}_3$  3 h, (i)  $\text{TbH}_3$  10 h.**



**Figure 5. Summary of EDX compositional profiles of (a)  $\text{DyH}_3$  and (b)  $\text{TbH}_3$**

## CONCLUSION

The heat treatment of heavy rare earth (Dy and Tb) hydrides provides an effective means of increasing the coercivity of sintered NdFeB-type magnets, but might be limited to the particular thickness of the sintered magnets. Such a process results in a slight drop in the remanence when compared to the conventional powder blending process. During the heat treatment, the Dy/Tb element diffused into the magnet probably only into the grain boundary, and form heavy rare earth-rich shells around the grain boundary of the  $\text{Nd}_2\text{Fe}_{14}\text{B}$  phase, which is similar to results on coating with Dy or Tb oxides and fluorides (15). It can be seen from Figure 5, that much amount of diffused Tb was observed than Dy, at depth of 60  $\mu\text{m}$  which is due to the difference in chemical reaction during heat treatment. This type process can provide an alternative route to modify the composition after sintering which can give a significant freedom throughout manufacturing to tailor magnetic properties.

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