Proceedings of the 2nd International Congress APMAS2012, April 26–29, 2012, Antalya, Turkey

Investigation of Grain Boundary Compositions and Magnetic Properties of Hot-Worked $Nd_{18}Tb_1Fe_{66.5}Co_5Al_{1.5}B_8$ Magnets

G. Sadullahoğlu[a], B. Altuncevahir[b] and A. Okan Addemir[a]

[a]Istanbul Technical University, Metallurgical and Materials Engineering, 34469, Maslak, Istanbul, Turkey

[b]Istanbul Technical University, Physics Engineering, 34469 Maslak, Istanbul, Turkey

Magnetic properties which are related to the compositions on the grain boundaries of the NdFeB permanent magnets were studied at different hot working temperatures applied to the cast $Nd_{18}Tb_1Fe_{66.5}Co_5Al_{1.5}B_8$ ingots. Since the addition of Al caused the formation of hard magnetic μ phase between the magnetic NdFeB crystals after the pressing 800 °C H_c value increased to the highest value of 8.21 kOe. According to the SEM micrographs and EDS analysis, the increase in pressing temperature, the atomic ratios in the NdFeAl compositions approached to hard magnetic μ phase with the increase in pressing temperature and its exact composition was obtained in the sample hot deformed at 800 °C. Although the secondary μ phase with a high melting temperature is detremental to the alignment of c axis of the magnetic NdFeB grains along the pressing direction, we found that the magnetic properties of the magnets improve due to the presence of this hard magnetic phase on the grain boundaries.

DOI: 10.12693/APhysPolA.123.233

PACS: 75.50.Ww, 75.50.Vv, 75.50.Bb

1. Introduction

Hot deformation of cast alloys is an alternative method for sintered high performance RE–Fe–B magnets. Magnetic alignment of the hard $Nd_2Fe_{14}B$ phase is achiEved by hot pressing, upset forging, hot rolling or extrusion [1–4]. The addition of Al to NdFeB magnets enhanced strongly H_c due to the increased liquid state wettability during sintering heat treatment and the formation of new phases stabilized by Al. In Fe–Nd–Al ternary system, several binary or ternary phases are in equilibrIum. Both δ ($Nd_{33}Fe_{70-x}Al_x$; $7 < x < 25$) and μ ($Nd_{33}Fe_{67-x}Al_x$; $2.5 < x < 5$) phases are stable on the Al poor part of the Nd–Fe–Al system and are observed in Fe–Nd–B–Al magnets [5–8]. In this study, we investigated the hot working temperature on the microstructure and magnetic properties of the Al doped NdFeB cast alloys.

2. Experimental

The alloy composition of $Nd_{18}Tb_1Fe_{68.5}Co_5B_8Al_{1.5}$ was prepared by induction melting in a quartz tube under vacuum about 10^{-3} mbar and solidified in water to achieve fine microstructure. Hot deformed magnets were produced by hot-pressing of the as-cast ingots in an open die at 710, 750 and 800 °C under high purity argon atmosphere. The pressure during the hot pressing was 2.55 ton/cm^2. Hot pressed samples were heat treated at 580 °C for 1 h. Scanning electron microscope (SEM) equipped by an energy dispersive spectrometer (EDS) was utilisied to investigate microstructure of the specimens. $2 \times 2 \times 2$ mm^3 specimens were cut from the hot pressed magnets. Magnetic measurements of the cubic samples were performed under a maximum magnetizing field of 20 kOe with a vibrating sample magnetometer (VSM). Thermal differential analyzing (DTA) was used to define phase transformations.

3. Results and discussions

The SEM micrographs of the as-cast ingot and hot pressed cast alloys at 710, 750 and 800 °C are shown in Fig. 1. The microstructure of the cast ingot consists mainly of two phases; $Nd_2Fe_{14}B$ ferromagnetic matrix phase (region (a)) and Nd-rich intergranular phase (region (b)). In Fig. 1a and b, the magnetic grains are isotropic and columnar shaped and no free iron was detected in the cast ingots in EDS analysis. According to the EDS results small part of Al entered the magnetic $Nd_2Fe_{14}B$ structure, the rest of Al remained in the intergranular region.

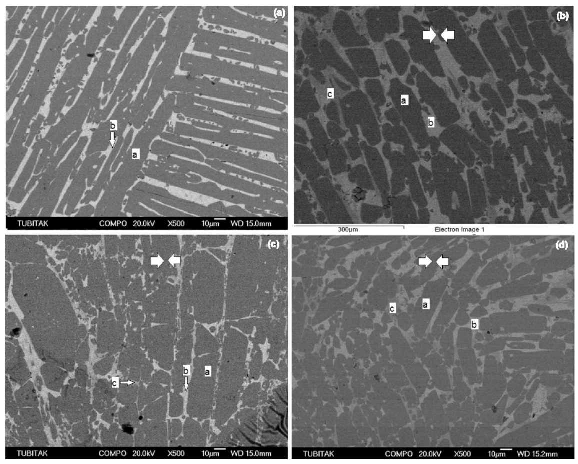

Fig. 1. SEM images of the samples as cast (a), hot pressed at 710 °C (b), 750 °C (c), and 800 °C (d). a: $Nd_2Fe_{14}B$, b: Nd-rich phase and c: FeNdAl composition. Arrows show pressing directions.

The intergranular regions were separated into two compositions after hot pressing process consisting of Nd-rich (a) and NdFeAl ternary composition (c).

TABLE

Atomic percents of the elements in region c in the hot pressed samples.

Pressing temp. [°C]	Atomic percent [%]				
	Fe	Nd	Co	Al	O
710	40.25	44.29	4.33	3.71	7.41
750	45.38	39.72	5.24	3.98	5.68
800	59.33	30.72	–	3.94	6.02

According to Table the amount of Nd in the NdFeAl phase decreased when the deformation temperature increased from 710 °C to 800 °C and the composition of μ phase was observed in the microstructure of the sample hot pressed at 800 °C. μ phase ($Nd_{33}Fe_{67-x}Al_x$; $2.5 < x < 5$) is hard ferromagnetic with a high anisotropy field ($\mu_0 H_A > 8$ T) and formed by peritectic reactions at about 750 °C [8]. The surface oxidation most probably occurred during preparation process of the samples for SEM analysis.

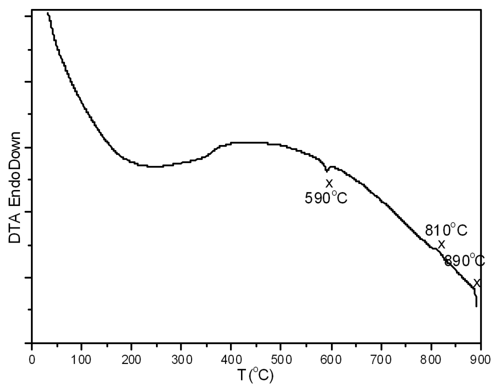

Fig. 2. DTA thermograph of the sample hot pressed at 800 °C.

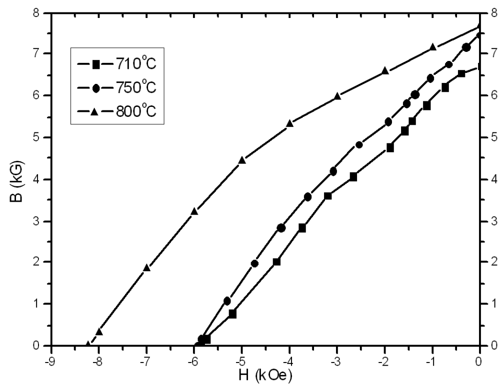

Fig. 3. Demagnetization curves of the samples hot pressed at different temperatures.

In the thermal analysis of the sample hot pressed at 800 °C (Fig. 2) the endothermic reaction occurred at about 590 °C which corresponds to melting temperature of Nd-rich phase in the intergranular region. The other exothermic and endothermic reactions observed at about 810 °C and 890 °C are attributed to $L + \delta + \psi \leftrightarrow \delta + \psi_2$ and $\delta + \psi_2 \leftrightarrow L + \psi_1 + \delta$ transformations, respectively, because μ phase disappears and those two reactions occur at 805 °C and 900 °C, respectively ($\delta = Nd_{30}Fe_{62-x}Al_{8+x}$ is tetragonal and antiferromagnetic, ψ, ψ_1 and ψ_2 phases are $Nd_2(Fe+Al)_{17}$ where Al and Fe ratios change).

Figure 3 indicates the demagnetization curves of the samples hot pressed at 710, 750, and 800 °C and annealed at 580 °C for 1 h. According to the demagnetization curves, the sample hot deformed at 800 °C has the highest B_r and H_c values which can be attributed to the formation of μ phase on the grain boundaries because it is a hard ferromagnetic and has large anisotropy field (H_A) which increase the coercivity significantly. B_r and H_c values increased to 7.67 kG and 8.21 kOe, respectively.

4. Conclusions

In this study, we obtained the highest B_r and H_c values for the sample hot deformed at 800 °C due to the presence of hard ferromagnetic μ phase on the grain boundaries. In the microstructures of the pressed samples the grain alignment is not good, because the high melting NdFeAl composition inhibits the grain rotation during hot pressing process. The improvement of the magnetic properties are most probably due to the hard magnetic properties of μ phase. In the microstructure of the cast alloy the intergranular region composes of only Nd-rich phase. After hot working process NdFeAl ternary compositions were observed in all samples. With the increase in pressing temperature, atomic ratios in the NdFeAl compositions approached hard magnetic μ phase and its exact composition was obtained in the sample hot deformed at 800 °C. The improvement of the magnetic properties of the hot pressed samples by the increase in deformation temperature might be attributed to the enhancement of the amount of μ phase in the microstructure by increasing pressing temperature.

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