A STUDY OF MAGNETIC ALIGNMENT OF DIE-UPSET Pr-Fe-B-Cu MAGNETS

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Abstract- An attempt has been made to investigate the mechanism of magnetic alignment in the magnets produced by upset forging the $Pr_{20}Fe_{74}B_4Cu_2$ cast bulk alloy. Upset forging of the cast alloy was carried out for 20 sec to an 80 % thickness reduction (strain rate: $4x10^{-2}s^{-1}$) in an open die configuration at varying temperatures in the range 600° - $900^{\circ}C$. It has been found that the upset forging process at temperatures above $800^{\circ}C$ can achieve a magnetic alignment to a great extent from copper-containing Pr-Fe-B-type cast ingot. The growth manner of the ferromagnetic $Pr_{2}Fe_{14}B$ matrix grain in Pr-Fe-B-type alloys was studied by examining the morphology change of the matrix grain in sintered body, and it was found that the matrix grains grew in anisotropic manner such that the grain grew more rapidly along the a- or b-axis than along the c-axis. This anisotropic grain growth led to the plate-like shape of the matrix grain. The magnetic alignment during the upset forging was attributed to grain boundary gliding of the plate-like grains, and the geometry of the grains in the cast ingot and the presence of a large amount of the praseodymium-rich grain boundary phase were thought to play a key role in the achievement of magnetic alignment.

L INTRODUCTION

The hard magnetic properties of the alloys based on RE₂Fe₁₄B compounds are generally poor in the cast state. Recently, however, it has been found that the Pr-Fe-B-type alloys can exhibit appreciable hard magnetic properties even in the bulk ingot state by employing a proper heat treatment and the minor addition of elements such as copper, aluminium and silver[1-5]. More interestingly, the copper-containing Pr-Fe-B alloys have been known to be successfully fabricated into high performance permanent magnets by upset forging at high temperature directly from cast ingot [6,7]. During the upset forging, a significant degree of magnetic alignment of the ferromagnetic Pr₂Fe₁₄B matrix grains along the forging direction is achieved. In the present study, an attempt has been made to study the mechanism of magnetic alignment during upset forging of the cast bulk Pr-Fe-B-type alloy.

II. EXPERIMENTALS

An alloy with a chemical composition Pr₂₀Fe₇₄B₄Cu₂ was melted using an induction furnace, and cast into a 7 mm thick water-cooled copper mould. 7x7x10 mm³ blocks were cut from the cast ingot so that the 10 mm longitudinal axis of the cut block was perpendicular to the cooling direction of the ingot. Upset forging of the blocks was carried out by pressing blocks along the longitudinal direction in order to take an advantage of the presence of preferred orientation (about the preferred orientation, see ref. [4]) for 20 sec to an 80 % thickness reduction (strain rate: 4x10⁻²s⁻¹) in an open die configuration at varying

temperatures in the range 600° - 900°C under argon gas atmosphere. Fig. 1 shows the schematic diagram of the pressing rig used in the present study. The magnetic domain structure of the samples was observed using a Kerr image method in an optical microscope. 3x3x1.5 mm³ specimens of the upset forged magnets were prepared for magnetic measurement using a VSM. The 1.5 mm thickness direction was parallel to the upset forging direction, and the measurements were taken along the upset forging direction. In order to investigate the growing manner of Pr₂Fe₁₄B matrix grain in Pr-Fe-B-type alloys, the morphology change of the matrix grain in pre-aligned sintered body produced using an equi-axed fine powder during sintering and subsequent annealing was studied. Pre-aligned and isostatically pressed compacts were sintered at 1050 °C for 1 hr and then annealed at 1000 °C for 24 hrs for growing the grains.

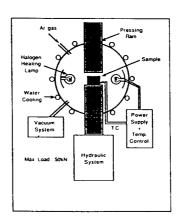


Fig.1. Schematic diagram of the upset forging rig.

III. RESULTS AND DISCUSSION

Fig.2 shows the variations of remanence of the upset forged magnets produced from the as-cast ingot as a function of forging temperature. The remanence of the upset forged samples increases with increasing upset forging temperature, and it appears that reasonably good remanence (Br: ~10.0 kG) can be achieved by upset forging at temperatures above 800°C. The good remanence of the magnets upset forged above 800 °C indicates that the magnetic matrix grains are well aligned during the upset forging process. The typical magnetic domain structure of the specimen forged at 900 °C observed on the plane perpendicular to the upset forging direction is shown in Fig.3, and it can be seen that the easy magnetisation direction (EMD : c-axis) of the ferromagnetic Pr₂Fe₁₄B matrix grains is well aligned along the upset forging direction.

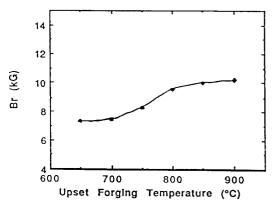


Fig.2. Variation of the remanence of the upset forged magnets as a function of upset forging temperature.

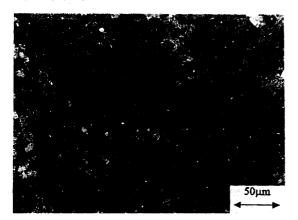


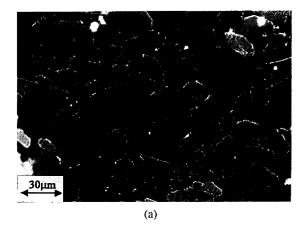
Fig.3. Magnetic domain structure (Kerr image) of the magnets upset forged at 900 °C (observed on the plane perpendicular to the forging direction).

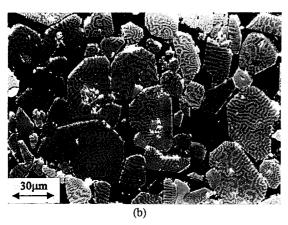
The microstructural and the magnetic measurement studies indicated that a significant easy axis magnetic alignment along the forging direction has been achieved during the upset-forging. This magnetic alignment can be explained by the grain boundary gliding of the plate-like matrix grains. As shown in Fig.4, most of the grains in the cast ingot have a plate-like shape and the EMD is roughly perpendicular to the flat surface of these grains. If such plate-like grains are squeezed at high temperature during upset-forging, they should reorientate so that the wide flat surfaces become perpendicular to the upset-forging direction. This alignment may take place through the grain boundary gliding process, and the praseodymium-rich phase, which is liquid at the upset-forging temperature (see Fig.7), would facilitate this process.



Fig. 4. Micrograph showing the morphology and the magnetic domain image of the grains in cast ingot.

Fig. 5 shows the microstructure and the magnetic domain structure of the sample (65 % thickness reduction) upset forged at 900 °C and then annealed for 2 hrs at 1000 °C. Fig. 5 (a) and (b) represent the magnetic domain image observed on the planes parallel and perpendicular to the upset forging direction respectively, showing that the EMD of the plate-like grains has been aligned along the upset forging direction. A further squeezing of the sample should produce further alignment. This microstructural observation clearly indicates that the magnetic alignment during upset forging takes place through the grain boundary gliding process. Fig.6 represents schematically the model for the achievement of magnetic alignment during upset forging.





Lig. 5. Kerr images showing the microstructure and the magnetic domain structure of the upset forged magnets (65% thickness reduction), observed on the planes parallel (a) or perpendicular (b) to the upset forging direction.

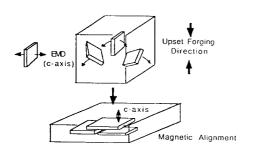


Fig.6. Schematic diagram showing the model for magnetic alignment in the upset forged magnets.

Fig.7 shows the DTA result for the Pr₂₀Fe₇₄B₄Cu₂ alloy. The result for copper-free Pr₂₀Fe₇₆B₄ alloy is also included in the Fig.7. On heating, there are two endothermic peaks for both alloys. The peak appeared at lower temperature may be corresponding to the Curie temperature of the Pr₂Fe₁₄B matrix grain, and the peak at higher temperature to the eutectic temperature of Pr-rich grain boundary phase. It is worth noting that the eutectic temperature of the grain boundary phase is significantly lowered from around 660 °C to around 460 °C by the addition of copper. The lowered eutectic temperature of the grain boundary phase suggests that the grain boundary gliding is further facilitated for the copper-containing alloys.

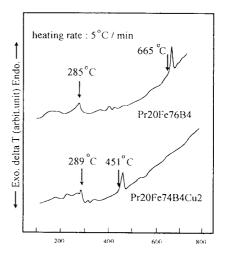


Fig.7. DTA results for the Cu-containing or Cu-free Pr-Fe-B alloys.

As discussed above, the presence of plate-like morphology of the Pr₂Fe₁₄B matrix grain was found to be a prerequisite for achieving the magnetic alignment of the matrix grain during upset forging of the cast ingot. The plate-like morphology of the grains in the ingot may be resulting from an anisotropic growth of the grain during solidification. This anisotropic growth of the matrix grain in Pr-Fe-B-type alloy was studied using analogy from morphology change of the matrix grain during sintering of Pr-Fe-B-type powder. Roughly equi-axed particles (see Fig.8) were sintered at 1050 °C and then annealed for 24 hours, and the morphology of grains in the sintered body was examined in detail by observing the morphology and the magnetic domain structure along different directions using a Kerr image method, and the results are shown in Fig.9.



Fig. 8. SEM micrograph showing the morphology of initial particles.

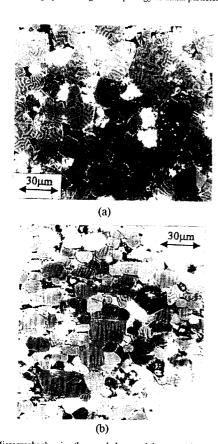


Fig.9. Micrographs showing the morphology and the magnetic domain image of grains in sintered body (Pr20Fe74B4Cu2 alloy) observed on the plane perpendicular (a) or parallel (b) to the aligning direction.

Fig. 9 (a) shows the morphology and the magnetic domain image of grains observed on the plane perpendicular to the aligning direction. The grains appear to be roughly circular and the axial ratio, A/B (see Table 1), of the grains measured on this plane was found to be very close to unity. Meanwhile, Fig 9 (b) shows the morphology of the grains observed on the plane parallel to the aligning direction. The grains appear to be elongated along the direction perpendicular to the aligning direction, and the axial ratio, A/C (see Table 1), of the grains measured on this plane was found to be around 1.81. These results indicate that the initial spherical particles is grown in an anisotropic manner during the sintering and subsequent annealing in such a way that the growth rate along the directions within the basal plane of the tetragonal Pr₂Fe₁₄B grain is higher that that along the c-axis direction of the grain. In Table 1, the axial ratios representing morphology of the Pr₂Fe₁₄B matrix grains in sintered body of the Pr-Fe-B-type alloys containing varying amount of copper are tabulated. It appears that the addition of copper to the Pr-Fe-B-type alloys enhance the anisotropic grain growth of the matrix grain. These results may explain the fact that better permanent magnetic properties are achieved from upset forged magnets produced using copper-containing alloys rather than copper-free alloys [1].

The grain growth during sintering and subsequent annealing may take place through a so-called 'dissolution and precipitation' process. The anisotropic grain growth in sintered body observed in the present study may be meant to show, by analogy, how the plate-like grains in the cast ingot of Pr-Fe-B-type alloys develops during solidification. More detailed study on the anisotropic growth of the Pr₂Fe₁₄B matrix grain in Pr-Fe-B-type alloys is in progress.

Table 1. The axial ratios representing morphology of the matrix grain in sintered body of Pr20Fe76-xB4Cux allovs.

х	initial particles	grains in sintered body	
	A/A raito	A/B raito	A/C raito
0	1.01	1.01	1.56
1.0	1.02	0.98	1.60
2.0	1.01	1.02	1.81
	A C	B Control	c ‡ আiiii
		observed along the aligning direction	observed along the direction perpendicular to the aligning direction

IV. CONCLUSIONS

It has been found that the upset forging process at temperatures above 800 °C can achieve a magnetic alignment to a great extent from copper-containing Pr-Fe-B-type cast ingot. The ferromagnetic Pr₂Fe₁₄B matrix grain in Pr-Fe-B-type alloys was found to grow in anisotropic manner such that the grain grew more rapidly along the a-or b-axis than along the c-axis, and this anisotropic grain growth led to the plate-like shape of the grain. The magnetic alignment during the upset forging was attributed to grain boundary gliding of the plate-like grains, and the geometry of the grains in the cast ingot and the presence of a large amount of the praseodymium-rich grain boundary phase were thought to play an important role in the achievement of magnetic alignment.

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