Journal of Alloys and Compounds, 189 (1992) 131–137
JALCOM 372

Study of Pr–Fe–B–Cu permanent magnets produced by upset forging of cast ingot

H. W. Kwon, P. Bowen and I. R. Harris
School of Metallurgy and Materials, The University of Birmingham, P.O. Box 363, Birmingham, B15 2TT (UK)

(Received May 18, 1992)

Abstract

An attempt was made to produce anisotropic Pr–Fe–B–Cu-type permanent magnets from the cast ingot materials using an upset forging process. The effects of the casting condition of the ingot on the magnetic properties of the upset forged magnets were investigated. Most of the free iron which existed in the cast ingot material was removed by upset forging at high temperatures (approximately 900 °C), and this was achieved by a solid–liquid peritectic reaction between the free iron and the praseodymium-rich grain boundary phase thought to be aided by cracks in the $Pr_2Fe_{14}B$ matrix grains caused by the upset forging. The magnetic alignment of the upset forged magnets can be explained by grain boundary gliding of the plate-like ferromagnetic matrix grains. A Pr–Fe–B–Cu magnet (${}_iH_c \sim 12.0$ kOe, $B_r \sim 10.5$ kG) with good demagnetization character was produced by the upset forging of the ingot material at 900 °C.

1. Introduction

Copper-containing Pr–Fe–B alloys with high praseodymium content (approximately 17 at.%) are known to show appreciable hard magnetic properties in the cast state, unlike the corresponding neodymium-based alloys [1–5]. The praseodymium-based alloys have also been successfully fabricated into high performance permanent magnets by hot pressing [6, 7]. The present authors have reported [3, 4] that, in the cast state, Pr–Fe–B–Cu with higher praseodymium content (20.5 at.%) exhibited better hard magnetic properties than the corresponding alloy with lower praseodymium content (17.5 at.%) and the alloy has been found to show some preferred orientation in the bulk ingot. The easy magnetization direction (EMD, the *c*-axis) of the $Pr_2Fe_{14}B$ matrix grain in the cast ingot appeared to be in a preferred direction perpendicular to the cooling direction.

In the present study, we attempted to produce anisotropic Pr–Fe–B–Cu-type permanent magnets by an upset forging process where, typically, deformation was carried out in approximately 20 s using a Pr–Fe–B–Cu alloy with high praseodymium content. The effects of the upset forging temperature and the casting condition of the ingot material on the magnetic properties of the upset forged magnets were investigated. The microstructures of the upset forged magnets and their relation to the magnetic properties were also studied and a possible mechanism for the achievement of magnetic alignment is proposed.

2. Experimental details

An alloy with a nominal composition $Pr_{20}Fe_{74}B_4Cu_2$ was melted using an induction furnace, and cast ingots were produced under two different casting conditions: (1) casting into a 7 mm thick water-cooled copper mould (7 mm thick ingot); (2) casting into a 30 mm thick iron mould (30 mm thick ingot). $7\times7\times10$ mm^3 blocks were cut from the ingots so that the longitudinal axis of the sample was perpendicular to the cooling direction of the ingot. Upset forging of the blocks was carried out by pressing them along this longitudinal direction for 20 s to an 80% thickness reduction (strain rate 4×10^{-2} s^{-1}) in an open die configuration at varying temperatures in the range 600–900 °C under argon gas and then holding the upset forged samples at the upset forging temperature for 5 min. Figure 1 shows schematically the pressing rig used in the present study. The maximum load available from the rig is 250 kN but the current work employed a load cell of 50 kN, and the samples were heated by four halogen lamps. The microstructures of the cast ingots and the upset forged magnets were observed using an optical microscope and scanning electron microscopy (SEM), and the magnetic domain structure of the samples was observed using a Kerr image method in an optical

0925-8388/92/$5.00

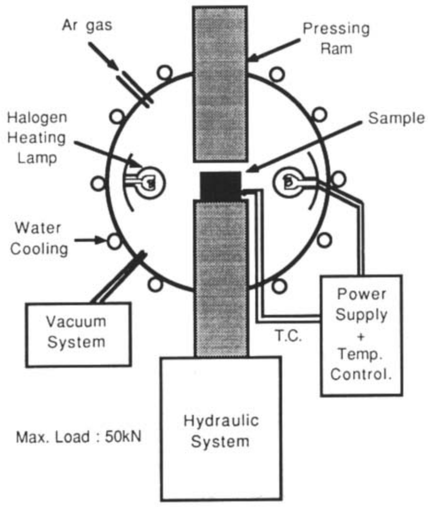


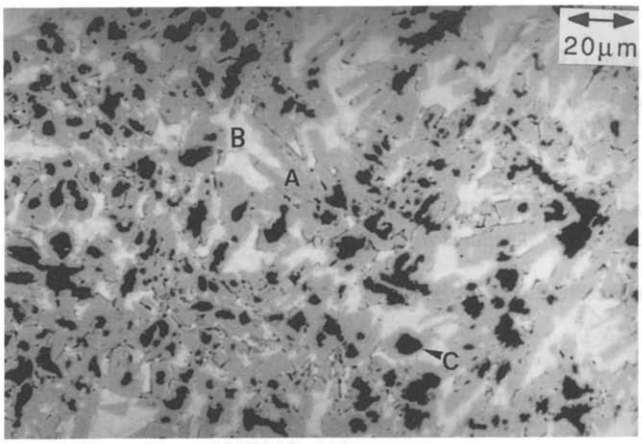
Fig. 1. Schematic diagram of the upset forging rig.

microscope. The grain sizes of the cast ingot were measured using an image analyzer. $3\times3\times1.5$ mm^3 specimens (approximately 100 mg) of the upset forged magnets were prepared for magnetic measurements (1.5 mm thickness direction parallel to the upset forging direction). Prior to the measurements, the specimens were magnetized along the upset forging direction using a magnetic pulser with a field strength of 4800 kA m^{-1}, and the measurements were taken along the upset forging direction. The permanent magnetic properties of the specimens were obtained from the second quadrant demagnetization curve.

3. Results and discussion

Figure 2 shows the microstructures of the 7 mm thick and the 30 mm thick ingots. The microstructures of both ingots consist mainly of three phases: a $Pr_2Fe_{14}B$ ferromagnetic matrix phase (A), a praseodymium-rich grain boundary phase (B), and free iron (C). A detailed study on the microstructure of the alloy used in the present study has been reported elsewhere [3, 4]. The obvious difference in the microstructure of the two ingots is the grain size of the matrix phase. The matrix grain size of the fast cooled ingot (7 mm thick) is much finer than that of the slow cooled ingot (30 mm thick). The matrix grain sizes of the ingots measured using an image analyser are shown in Table 1. These two ingot materials with different grain sizes were used for production of the upset forged magnets, and the effect of the grain size of the ingot material on the magnetic properties of the upset forged magnet was investigated.

Figure 3 is a typical micrograph showing the grain morphology and the magnetic domain structure of the ingot material (30 mm thick); it can be seen that most of the grains have a plate-like shape and the easy



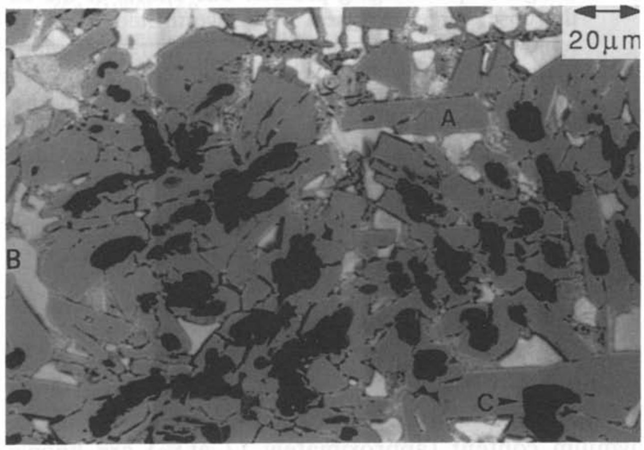


Fig. 2. SEM (backscattered electron) images showing microstructures of the cast materials: (a) 7 mm thick ingot, (b) 30 mm thick ingot.

TABLE 1. Mean matrix grain size of the ingots

Ingot	7 mm thick	30 mm thick
Grain size (μm)	≈ 10.5	≈ 18.0

magnetization direction (EMD, the c-axis of the grain) is roughly perpendicular to the flat surface of these grains.

Figure 4 shows the variations of the intrinsic coercivity and the remanence of the upset forged magnets produced from the 7 mm thick ingot as a function of the upset forging temperature. The intrinsic coercivity and the remanence of the upset forged alloys increase with increasing upset forging temperature, and it is found that reasonably good properties (${}_iH_c \approx 8.0$ kOe, $B_r \approx 10.0$ kG) could 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

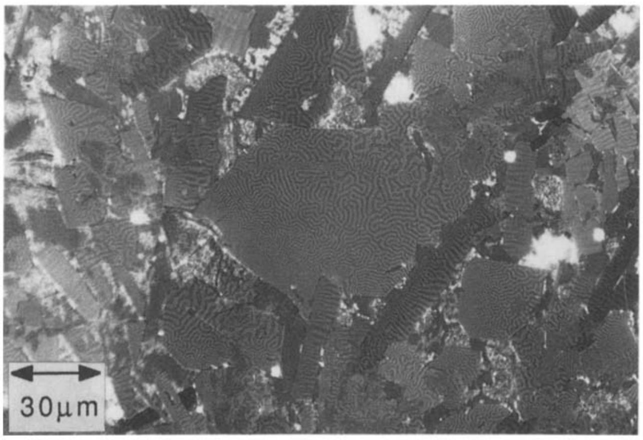


Fig. 3. Micrograph showing the grain morphology and magnetic domain structure of cast material (30 mm thick ingot).

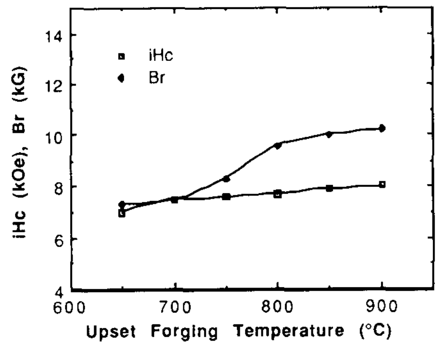


Fig. 4. Variations in the intrinsic coercivity ${}_iH_c$ and the remanence B_r of the upset forged magnets as a function of the upset forging temperature (7 mm thick ingot).

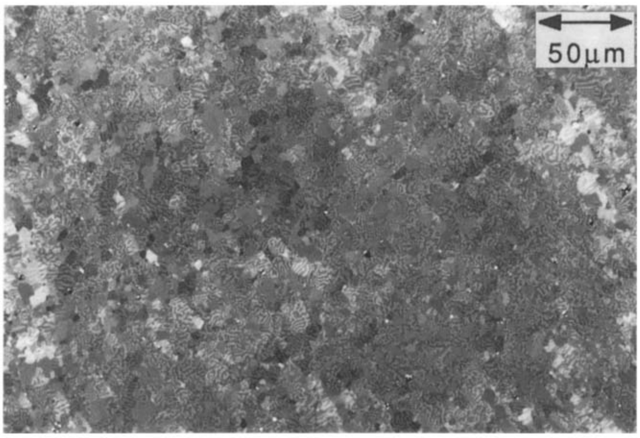
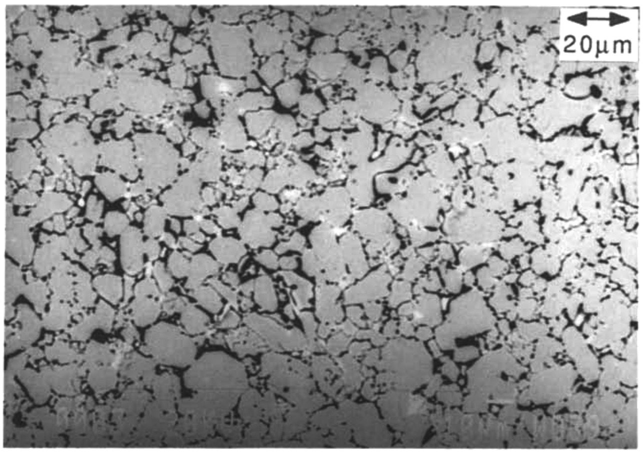


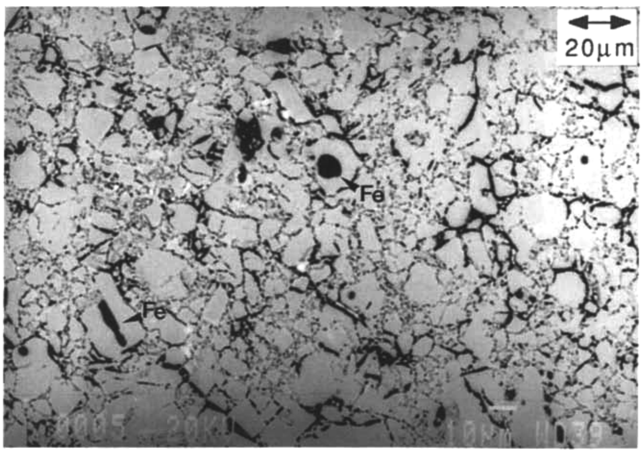
Fig. 5. Magnetic domain structure (Kerr image) of the magnet upset forged at 900 °C (7 mm thick ingot).

magnetic domain structure of the alloy upset forged at 900 °C observed on the plane perpendicular to the upset forging direction is shown in Fig. 5, and it can be seen that the EMD (c-axis) of the ferromagnetic matrix grains is well aligned along the upset forging direction. It should be interesting to compare the magnetic properties of upset forged magnets with those of sintered magnets. A sintered magnet with a composition very close to that used in the present study has been reported [8] to possess magnetic properties of $B_r = 10.7$ kG and ${}_iH_c = 19.8$ kOe. The intrinsic coercivity of the sintered magnet is significantly higher than that of the upset forged magnet, and this can be attributed to the fine grain size of the sintered magnet.

There is a striking change in the microstructure of the magnets upset forged at temperatures above 800 °C. Most of the free iron which existed in the initial ingot (Fig. 2) was removed by the upset forging process (see Fig. 6(a)). At first sight this is rather surprising since the simple upset forging process consisted of a short period of pressing (20 s) and a dwell time of only 5 min at the upset forging temperature. It has been reported [3, 4] that a long (over 5 h) annealing at 1000 °C is required to remove the free iron from the cast alloy used in the present study. Free iron is



(a)



(b)

Fig. 6. SEM (backscattered electron) images showing the microstructures of the magnets (30 mm thick ingot) upset forged at (a) 900 °C and (b) 750 °C.

still visible in the magnets upset forged at temperatures lower than 750 °C (see Fig. 6(b)).

The rapid removal of free iron by upset pressing at high temperature can be explained in the following manner. During upset forging, the $Pr_2Fe_{14}B$ matrix grain will be heavily cracked (see Fig. 7). The praseodymium-rich grain boundary phase, which will be liquid at the upset forging temperature, can then penetrate into the matrix grain through the cracks to make direct contact with the free iron. A peritectic reaction between the free iron and the praseodymium-rich liquid phase can, therefore, easily take place at the high temperatures, thus forming $Pr_2Fe_{14}B$ at the expense of the free iron and the praseodymium-rich phase.

The upset forged magnets were subjected to a post-upset annealing at 1000 °C, and the variations in intrinsic coercivity and remanence of the magnets upset forged at 900 °C as a function of annealing time for the 7 mm thick and 30 mm thick ingots are shown in Fig. 8. The magnetic properties of the magnets appeared to be improved slightly by annealing for 1–2 h, and prolonged annealing (over 6 h) appeared to deteriorate the properties. The overall values of the properties appeared to be similar between the upset forged magnets produced from the different ingots except for the slightly higher intrinsic coercivity for the 7 mm thick ingot. These results indicate that the magnetic properties of the upset forged magnets are not influenced appreciably by the casting condition of the ingot materials. It is also found that the post-upset annealing may not radically enhance the properties of the magnets upset forged at high temperatures. In Fig. 9, demagnetization curves of the upset forged and post-upset annealed magnets are shown.

For the magnets upset forged at the lower temperatures, however, the post-upset annealing was found to improve radically the intrinsic coercivity. In Fig. 10, the intrinsic coercivity and the remanence of the upset forged magnets produced from a 7 mm thick ingot after post-upset annealing at 1000 °C for 2 h are plotted against the upset forging temperature. The intrinsic coercivities of the magnets upset forged at the lower temperatures were improved radically by the post-upset annealing (compare with Fig. 4). The magnets upset forged at the lower temperatures will have a larger amount of grain boundary phase together with free iron. During post-upset annealing, the free iron will

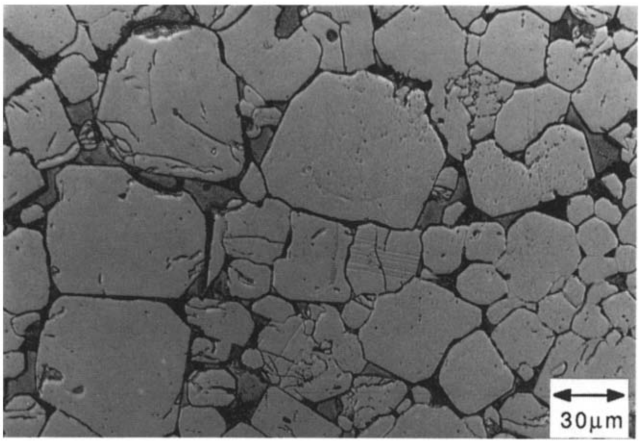


Fig. 7. Micrograph showing the transgranular cracks inside the matrix grains caused by upset forging (75% thickness reduction, 30 mm thick ingot).

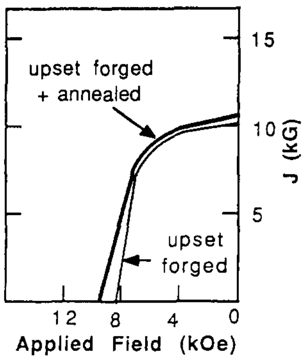


Fig. 9. Demagnetization curves of the magnets upset forged at 900 °C (post-upset annealing at 1000 °C for 3 h).

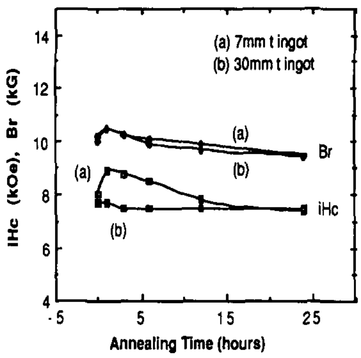


Fig. 8. Variations in intrinsic coercivity and remanence as a function of post-upset annealing time for the magnets upset forged at 900 °C.

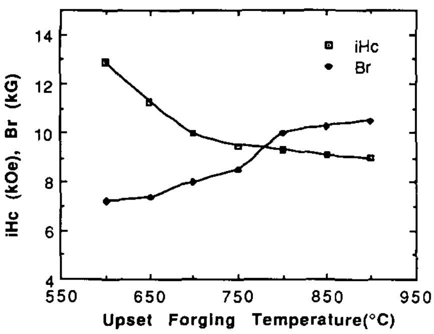


Fig. 10. Variations in intrinsic coercivity $_iH_c$ and remanence B_r as a function of upset forging temperature for the post-upset annealed (1000 °C for 2 h) magnets (7 mm thick ingot).

be removed and the praseodymium-rich grain boundary phase redistributed uniformly throughout the grain boundaries. The presence of more grain boundary phase would then result in better magnetic isolation between the $Pr_2Fe_{14}B$ magnetic matrix grains, thus enhancing the intrinsic coercivity of the magnets. In addition to the better magnetic isolation, the low internal demagnetizing field due to the poorer magnetic alignment [9, 10] may also contribute to the enhanced intrinsic coercivity of the magnets upset forged at the lower temperatures.

The intrinsic coercivities of the post-upset annealed magnets were improved further by a subsequent low temperature annealing. Figure 11 shows the variations in intrinsic coercivity and remanence of the post-upset annealed magnets (upset forged at 900 °C using a 7 mm thick ingot, post-upset annealed at 1000 °C for 2 h) as a function of annealing time at 500 °C. The intrinsic coercivity of the magnets was improved by around 3 kOe (9 kOe to 12 kOe) after the low temperature annealing. This improvement is probably related to the grain boundary modification caused by the low temperature annealing at 500 °C, which is just above the eutectic point of the grain boundary phase [4]. The previous study [3, 4] on the cast material showed that such a low temperature annealing treatment caused a significant improvement in the intrinsic coercivity, and this was found to be related closely to the formation of the $Pr(Fe,Cu)_2$-type phase at the grain boundary. No improvement in the remanence was achieved by the low temperature annealing treatment.

Figure 12 shows the second quadrant magnetization curve of the magnet upset forged at 900 °C. The magnet was produced from a 7 mm thick ingot and subjected to post-upset annealing at 1000 °C for 2 h and subsequent low temperature annealing at 500 °C for 4 h. The second quadrant magnetization curve for a sintered Nd–Fe–B-type commercial magnet (Philips, RES-270) measured under the same conditions as the upset forged

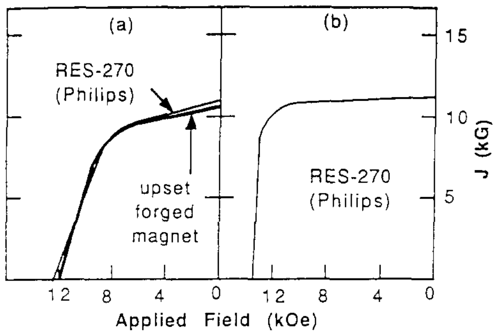


Fig. 12. Demagnetization curves of the upset forged magnet and the commercial sintered magnet measured by (a) VSM and (b) permeameter.

magnet is included for comparison. The demagnetization character of the upset forged magnet appeared to be comparable with that of the commercial magnet except for the slightly lower remanence of the upset forged magnet. It should be noted that these magnetic measurements using a VSM for samples with the shape $3\times3\times1.5$ mm^3 used in the present study will introduce a demagnetizing effect due to specimen geometry, and this would reduce the remanence value and result in a deterioration in the squareness of the loop. Figure 12(b) shows the demagnetization curve of the cylindrical commercial magnet ($\phi25\times10$ mm) measured using a permeameter, where the demagnetizing effect can be virtually eliminated. This curve shows a reasonably good squareness. It can be concluded, therefore, that the upset forged magnet exhibits demagnetization characteristics comparable with those of the Nd–Fe–B-type commercial sintered magnets.

As discussed above, the microstructural and the magnetic measurement studies indicate that the upset forging process is able to achieve a significant easy axis magnetic alignment along the upset forging direction. This magnetic alignment can be explained by the grain boundary gliding of the plate-like matrix grains. As shown in Fig. 3, 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, would facilitate this process, especially in an open die configuration. Figure 13 shows the microstructure and the magnetic domain structure of the upset forged sample (65% thickness reduction, 30 mm thick ingot) at 900 °C and then annealed for 2 h at 1000 °C. Figure 13(a) and (b) represent the images observed on the planes parallel and perpen-

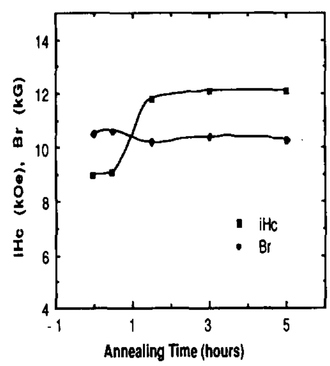
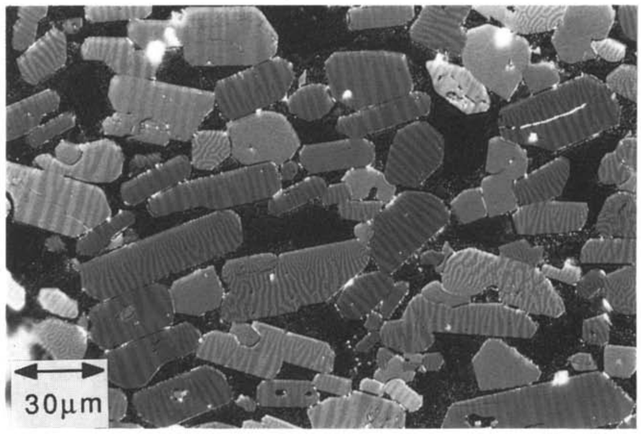
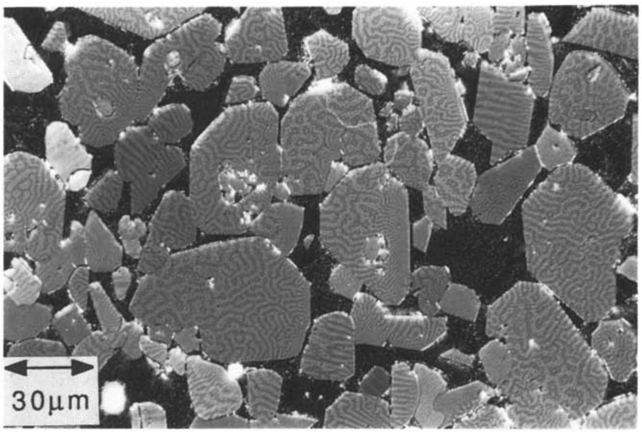


Fig. 11. Effect of low temperature (500 °C) annealing on the intrinsic coercivity ${}_iH_c$ and remanence B_r of the upset forged and then post-upset annealed magnets.



(a)



(b)

Fig. 13. Optical micrographs showing the microstructure and the magnetic domain structure of the upset forged magnet (65% thickness reduction) observed on the planes (a) parallel and (b) perpendicular to the upset forging direction (30 mm thick ingot).

dicular 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 and the grain boundary phase may be discharged. At the same time, the grains will be heavily deformed, and cracking of the matrix grain would take place (see Fig. 8). It seems, therefore, that the grain refinement occurs during the upset forging process by means of this cracking and then filling of the cracks by the liquid praseodymium-rich material. Figure 14 represents schematically the model for the achievement of magnetic alignment and the grain refinement discussed above.

It should be interesting to compare the magnetic alignment and grain refinement obtained in the upset forging process with those in the hot pressing process where the material is deformed with a fairly low strain rate (10^{-3}–10^{-5} s^{-1}). It has been reported [11] that dynamic recrystallization may play a vital role in the magnetic alignment and the grain refinement in the

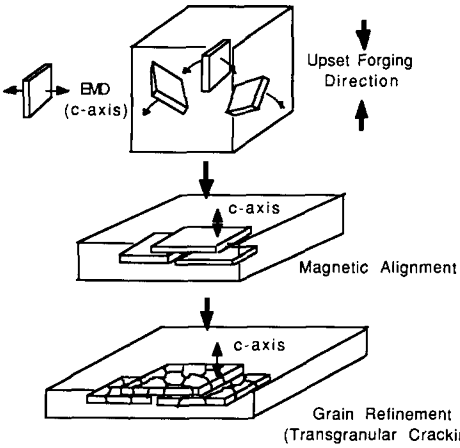


Fig. 14. Schematic diagram showing the model for magnetic alignment and grain refinement of the upset forged magnets.

hot pressing process. Other workers [12] have argued that the magnetic alignment in the hot pressing process may take place by slip of the matrix grain preferentially along basal planes. In the present upset forging process, however, the dynamic recrystallization may not allow reorientation since the upset forging is completed within a few minutes (about 5 min). The microstructural observation in the present study also showed clear evidence for the reorientation of the matrix grains. The magnetic alignment may, therefore, be achieved through this grain reorientation and there is no evidence of slip formation. The rapid achievement of preferred orientation obtained in the present work might have important implications with regard to the production of magnets directly from cast material.

4. Conclusion

It was found that the upset forging process at temperatures above 800 °C can be used successfully to produce Pr–Fe–B–Cu-type permanent magnets from the cast ingot materials with acceptable properties. It was found that the casting conditions of the ingot did not influence significantly the magnetic properties of the upset forged magnets. Most of the free iron which existed in the cast ingots was removed by the upset forging at high temperature, and this removal of the free iron was attributed to a solid–liquid peritectic reaction between the free iron and the praseodymium-rich grain boundary phase through the cracks in the matrix grains caused by the upset forging process. The magnetic alignment during the upset forging was attributed to grain boundary gliding of the plate-like grains, and the geometry of the ingot grain 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.

Acknowledgment

Thanks are due to the SERC and CEAM for the provision of research grants. Thanks are also due to R.E.P. (Widnes, UK) for the provision of alloys and to G. Mycock for valuable discussions.

References

1 T. Shimoda, K. Akioka, O. Kobayashi and T. Yamagami, *J. Appl. Phys., 64* (1988) 5290.
2 G. C. Hadjipanayis, M. Zhang and C. Gao, *Appl. Phys. Lett., 54* (1989) 1812.
3 H. W. Kwon, P. Bowen and I. R. Harris, *J. Appl. Phys. 70* (1991) 6357.
4 H. W. Kwon, P. Bowen and I. R. Harris, *J. Alloys Comp., 182* (1992) 233.
5 R. N. Faria, J. S. Abell and I. R. Harris, *J. Appl. Phys., 70* (1991) 6104.
6 T. Shimoda, K. Akioka, O. Kobayashi and T. Yamagami, *IEEE Trans. Magn., 25* (1989) 4099.
7 K. Akioka, O. Kobayashi, T. Yamagami and T. Shimoda, *J. Appl. Phys., 69* (1991) 5829.
8 R. N. Faria, J. S. Abell and I. R. Harris, *J. Alloys Comp., 177* (1991) 311.
9 D. Givord, P. Tenaud and T. Viadieu, in Mitchell, J. M. D. Coey, D. Givord, I. R. Harris and R. Hanisch (eds.), *Concerted European Action on Magnets,* Elsevier, London, 1989.
10 G. Martinek, D. Kohler and H. Kronmuller, Relations between the microstructure and magnetic properties of permanent magnets, *CEAM 2 Rep.,* September 1991 (Concerted European Action on Magnets), unpublished results.
11 T. Shimoda, Cast Pr–Fe–B magnet, *The Global Business and Technical Outlook for NdFeB Magnet Markets, 1989*, Gorham Advanced Materials Institute, CA, unpublished results.
12 Y. Luo and N. Zhang, *Proc. 10th Int. Workshop on Rare-Earth Magnets and Their Application, Kyoto, 1989,* p. 275.