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TEM Studies of the Microstructure and Domain Structure of Melt-Spun Iron-Rare Earth-Based Alloys

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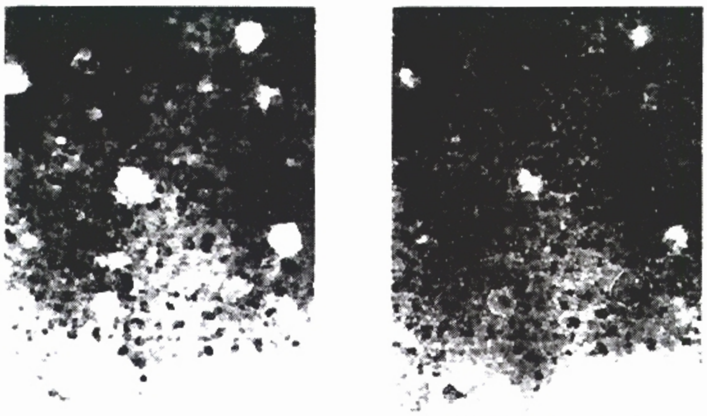
Alloys based on the NdFeB system are now well established as useful permanent magnets. However, a number of alloys based on other iron-rare earth systems have similar intrinsic magnetic properties to those of NdFeB permanent magnets, and it is important to continue to search for alternative systems. One such system is that based on the $Sm(Fe_{11}Ti)$ phase, which has the $ThMn_{12}$ structure, a high uniaxial anisotropy of 10.5T at room temperature, (somewhat higher than that of $Nd_2Fe_{14}B$), combined with a reasonably high saturation magnetisation. Specimens of composition $SmFe_{11}Ti$ were prepared by melt spinning in the form of amorphous ribbons, which were subsequently annealed to produce a small-grain polycrystalline material. Two series of heat treatments were carried out; in one series, specimens were annealed at 800°C for various times, and in the other series, specimens were annealed for 10 minutes at various temperatures. The first series of specimens were prepared by H Sun, Y Otani and J M D Coey (Trinity College, Dublin), and the second by J Ding (Ruhr-Universitat Bochum). The heat treatments and coercivities are summarised in the Table.



(a) In focus (b) Underfocussed

Fig. 1 TEM images of a $Sm(Fe_{11}Ti)$ specimen, annealed at 800°C for 30 mins

Specimen number	Anneal temp (°C)	Anneal time (min)	Hc(T)
1	800	10	0.49
2	800	20	0.56
3	800	30	0.37
4	650	10	0.14
5	750	10	0.36
6	825	10	0.54

It is seen that in the isothermal series of treatments (specimens 1-3), the coercivity H_c reaches a maximum at an anneal time of 20 minutes, whereas in the isochronal series (specimens 4-6), H_c increases with anneal temperature, and may not have reached its maximum value at 825°C.

In order to interpret these results, a transmission electron microscope (TEM) study of the specimens was carried out. It was found that specimens 1 and 2 consisted of polyhedral grains with diameters in the region of 50nm. The only phase detected was the $Sm(Fe_{11}Ti)$ phase. In specimen 3, some additional larger grains were found, which were identified as α-Fe by transmission electron diffraction and energy-dispersive X-ray analysis. Figures 1(a) and (b) show transmission electron micrographs of this specimen. In the out-of-focus micrograph (b), it may be seen that these larger grains are multi-domain, as shown for example by the grain near the centre, which contains a cross-shaped pattern of domain walls. Some α-Fe grains were also found in specimens 4-6, although they were less regularly distributed. However, it was found that in specimen 4, the grains were more angular with sharp corners, whereas in specimens 5 and 6, the grain boundaries were smoother.

These TEM studies have therefore helped to explain the variation of H_c with heat treatment. In the isothermal series, the decrease of H_c on over-annealing may be due to the presence of α-Fe grains, which can easily change their magnetisation direction, and may therefore act as nucleation centres for magnetisation reversal. In the isochronal series, annealing at low temperatures may produce grains with irregular-shaped boundaries. The large demagnetising fields produced by these grain shapes may lead to easy magnetisation reversal.

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Developments in Cast and Hot Worked PrFeB Magnets

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Introduction

Permanent magnets produced directly from the cast alloy would have the advantage of being a low-cost process of preparing rare-earth/transition-metal magnets. Two steps are needed in the processing: casting and annealing. The main difficulty in this process is to maintain a suitable alignment of the crystals during the solidification process. The production of magnets from cast material also has the distinct advantage of not involving a powder stage. If good magnetic properties can be achieved reproducibly by such a method then it should be possible to make significant savings on the production cost of magnets. Moreover, net shape magnets become a real possibility by hot working of cast material especially if shape forging at high strain rates can be tolerated, although it has been reported[1] that preferred orientation in Pr-Fe-B-Cu hot pressed magnets could be obtained only at very low strain rates. In hot-rolled magnets, it has been reported[2] that the final magnetic properties were dependent on the microstructure of the cast ingot. Thus, the solidification behaviour of the cast alloy can also be an important variable for hot worked magnets.

Cast Magnets

Crystallographic preferred orientation can be achieved by varying the temperature gradient/growth rate ratio (G/R) in the solidifying solid-liquid interface. This can be achieved in mould casting by changing such factors as the alloy composition, the temperature of pouring and the mould cooling characteristics (ie dimensions and material). In the case of dendritic growth ($Pr_2Fe_{14}B$ crystals as the matrix phase) appropriate values of G and R must be established for a particular alloy.

Figure 1 shows the demagnetisation curves for the annealed (1000°C, 24 hours) as-cast $Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ alloy cast in two different water cooled copper moulds. As can be seen, the solidification conditions have a dramatic effect on the remanence and intrinsic coercivity. The $_iH_c$ of the sample cast in the 30mm thick mould is much higher than that of the sample cast in the 7mm mould, and the latter exhibits a squarer loop with enhanced values of the B_r and $(BH)_{max}$. These observations are consistent with an increased c-axis alignment in the latter and the more isotropic nature of the former[3].

The effects of annealing at 1000°C and then furnace cooling was to remove the free iron from the matrix phase and to change the nature of the grain boundary phase. After annealing, the grain boundary region consisted of a coarser and more defined eutectic mixture (Pr/Cu-Pr) and also a sharper morphology of the Pr-rich phase[4].

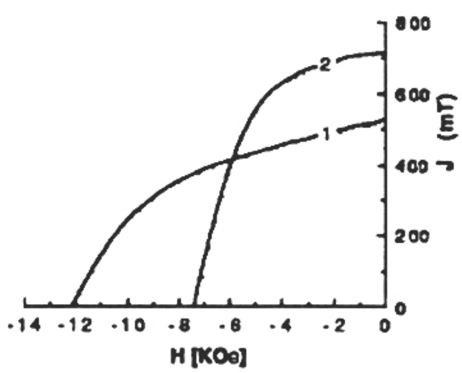

Fig. 1 Demagnetisation curves for PrFeBCu cast magnets. Curve 1 for 30mm thick mould and curve 2 - 7mm mould

Hot Worked Magnets

Better alignment of the $Pr_2Fe_{14}B$ crystals can only be obtained by mechanically working the cast ingot since in the cast state, the c-axis of the $Pr_2Fe_{14}B$ matrix phase is radially orientated perpendicular to the growth direction. An attempt has been made to produce anisotropic Pr-Fe-B-Cu-type permanent magnets from the cast materials ($Pr_{20.5}Fe_{73.8}B_{3.7}Cu_2$ alloy) by means of an upset forging process. Upset forging of the ingots was carried out by pressing them for 20 seconds to 80% thickness reduction (strain rate: 4×10^{-2} sec^{-1}) in an open die configuration at varying temperatures 600° - 900°C under Ar gas and then holding the upset forged samples at the forging temperature for 5 mins[5].

The $_iH_c$ and the B_r of the upset forged alloys appeared to increase with increasing upset forging temperature, and it was found that reasonably good properties could be achieved by upset forging at temperatures above 800°C. A magnet with good demagnetisation characteristics (see Figure 2) was produced by the upset forging of the ingot material at 900°C. Further annealing at 500°C led to an $_iH_c$ of ~12KOe (B_r ~10.5KG). The good remanence of the magnets indicated that the easy axis of the matrix grains was well aligned during the upset forging process along the forging direction.

This magnetic alignment could be explained by the grain boundary gliding of the plate-like matrix grain. Most of the grains in the cast ingot had a plate-like shape and the easy magnetisation direction (EMD) was roughly perpendicular to the flat surface of these grains. If such plate-like grains are squeezed at high temperature during the upset forging and are free to move then they should reorientate so that the wide flat surfaces

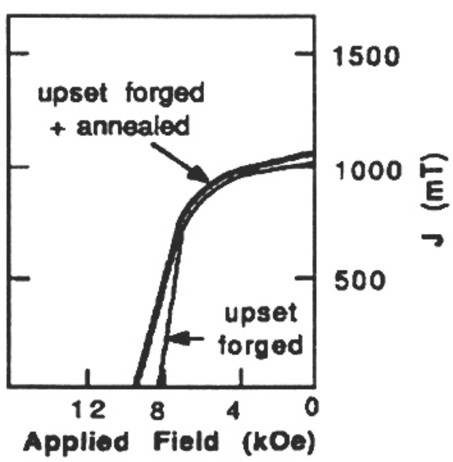

Fig.2 VSM demagnetisation curves of the magnets upset forged at 900°C (post-upset annealing at 1000°C for 3 hours

become perpendicular to the upset forging direction. The Pr-rich phase, which is liquid at the upset forging temperature, would facilitate this magnetic alignment process especially in an open die configuration (see Figure 3).

There was 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 was removed by the upset forging process. At first sight this is rather surprising since the simple upset forging

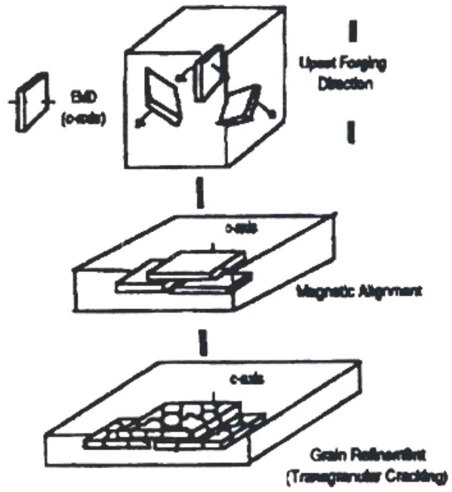

Fig.3 Schematic diagram showing the model for the magnetic alignment and grain refinement of the upset forged magnets

process consists of a short period of pressing (20 seconds) and a dwell time of only 5 mins at the upset forging temperature. However, the rapid removal of free iron by upset pressing at high temperature can be explained by the $Pr_2Fe_{14}B$ matrix grains becoming heavily cracked due forging. The Pr-rich grain boundary phase, which will be liquid at the upset forging temperature, then penetrates into the matrix grain through the cracks to make direct contact with the free iron. This would facilitate the peritectic reaction between the free iron and the Pr-rich liquid phase, thus forming a $Pr_2Fe_{14}B$ ferromagnetic phase at the expense of the free iron and the Pr-rich phase.

The overall values of the magnetic properties appeared to be similar between the upset forged magnets produced from the different ingots except for the slightly higher intrinsic coercivity for the 7mm thick ingot. This indicates that the magnetic properties of the upset forged magnets are not influenced appreciably by the casting condition of the ingot materials.

Conclusions

Significant differences in the magnetic properties of cast magnets can be obtained by casting in moulds with different cooling rates. Upset forged

concluded opposite