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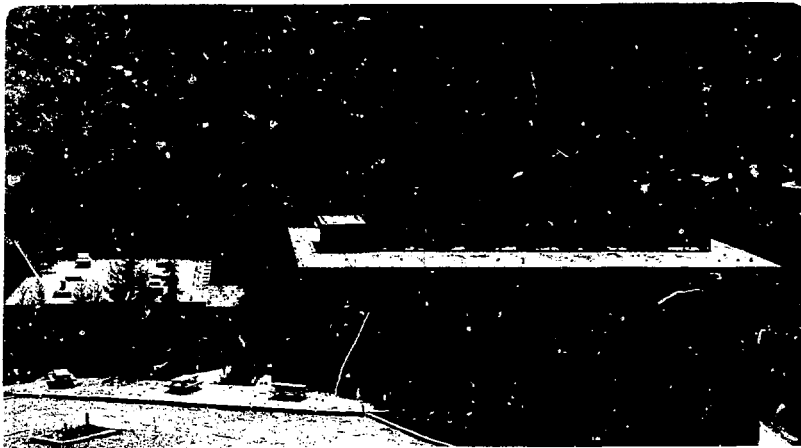
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MICROSTRUCTURE AND PROPERTIES OF STEP AGED RARE
EARTH ALLOY MAGNETS

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Microstructure and Properties of Step Aged Rare Earth Alloy Magnets

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ABSTRACT

Alloys with Compositions Co-25.5wt/o Sm-8w/o Cu-15 w/o Fe-3w/o Zr and Co-Sm-Cu-Fe-1.5 w/o Zr have been step aged to produce magnets with coercive force (iHc) in the range of 10-25kOe, much higher than those reported so far in the literature for the Zr alloys. The high coercive force magnets are typically aged at 800-850°C for 10-30 hours following the solution treatment at 1150°C. Subsequently, these are step aged to produce materials with high coercivity. The microstructure in all these alloys has a 2 phase cellular morphology with 2:17 phase surrounded by a 1:5 boundary phase. The long aging treatments at 800-850°C lead to coarsening of the two phase structure. The subsequent step-aging does not change the morphology, but only changes the chemical composition of the two phases.

Best properties are obtained in materials with a coherent microstructure of optimum boundary phase thickness and optimum chemical composition. The highest values of iHc obtained so far are ~ 26kOe and ~ 16 kOe for the 3% Zr and 1.5% Zr alloys respectively. The best hard magnetic properties of (B_H)_{max} = 33 MGOe and iHc = 13kOe are for a 25% Sm-20%Fe-4 Cu-2%Zr alloy.

INTRODUCTION

Efforts to design hard magnetic materials based on the Sm₂ (Co,Cu,Fe) system have led to the investigation of the effects of various heat-treatments and additions of other alloying elements on the magnetic properties. (1,2,3) Alloys containing small amounts of Zr have been shown to have energy products ~ 30 MGOe, (1) despite the low iHc (~7kOe). The microstructures of these alloys consist of a cellular morphology with 1:5 type phase surrounding the 2:17 "cells" (4) The

The magnetisation mechanisms in these materials has been shown to be the domain wall pinning mechanism (4). Although the role of Zr in these alloys is not yet well-understood, it is believed that the step aging treatments lead to chemical partitioning between the 1:5 and 2:17 phases, and thus harden the material magnetically, without any morphological changes occurring during step aging (4).

In this paper, the effects of the chemical compositions and heat treatments on the magnetic properties and the microstructure are studied systematically to establish the relationships between the microstructure and the properties on one hand and the microstructure and the chemistry, processing history, etc. on the other. It has been possible to prepare material with $(BH)_{max}$ of 33MG0e and iH_c of 16k0e for certain chemical compositions and heat treatments discussed below. The microstructural features responsible for high coercivity in this class of materials are also identified in the paper.

Mainly alloys of Co with 24-27wt%Sm, 4-8wt%, 15-20 wt%Fe and 1-3wt%Zr were studied. All alloys were prepared by melting together the component metals of samarium, cobalt, copper and iron, all with purities exceeding 99.5wt%, and ferrozirconium, in an induction furnace under argon atmosphere. The cast ingots were crushed and pulverized to 3-5 μ m particles with a jet mill. The powder thus obtained was pressed at a pressure of 1.5ton/cm² in a magnetic field of 10k0e. The green bodies were sintered for 1 hour at various temperatures between 1150 and 1250°C. These specimens were solution treated for 1 hour at a temperature between 1100 and 1220°C, then quenched in argon atmosphere. Following the solution treatment, they were aged by using isothermal aging and step aging. In this study, step aging process is as follows: following the isothermal aging, specimens were continuously cooled to 400°C at cooling rate of 1/2°C./min and subsequently aged at 400°C for 10 hours. The magnetic properties of these specimens were measured with an automatic recording flux meter. Part of them were magnetized by pulse magnetizer. Electron transparent specimens were prepared from thin platelets containing the C-axis in the plane of the disc, via ion milling. These foils were examined in a Philips EM 301 microscope operating at 100kv.

RESULTS AND INTERPRETATION

The effects of varying amounts of Sm, Fe, Cu and Zr on iH_c are shown in Figs. 1, 2 and 3 for both isothermally aged and step aged alloys. iH_c after isothermal aging decreases with increasing Sm content from 20kOe at 23 wt/o Sm to 2kOe at 25.5 wt/o Sm. However, step aging leads to an increase of iH_c for alloys with higher Sm content (fig. 1). Increasing amounts of Sm also improves the squareness of the demagnetisation curve.

While isothermal aging at 850°C for 1 hour does not have any effect on the iH_c for alloys containing varying amounts of Zr, step aging improves the iH_c markedly for these alloys; specifically for the alloys containing 2-3 wt/o Zr. For these alloys, the squareness of the demagnetisation curve also improves (fig. 2).

iH_c does not change with changing Fe concentration for isothermally aged alloys. However, step aging improves the iH_c and as can be seen in Fig. 3, depending on the Cu content in the alloy, iH_c attains a maximum after step aging.

The effect of Cu on iH_c for given Sm or Fe content in Figs. 1 or 3 is to have large iH_c for alloys with higher Cu concentration. In all these cases, the amount of improvement in iH_c after step aging depends on the specific composition of the alloy.

Fig. 4 shows the effect of isothermal aging time on iH_c for 1.5 wt/o Zr and 3 wt/o Zr alloys. Also, iH_c for alloys, which are aged isothermally for longer times (> 8 hours for 1.5 wt/o Zr alloys and > 3 hours for 3 wt/o Zr alloys) increases rapidly after step aging (fig. 4) up to a maximum value of ~26kOe.

The microstructures of all these alloys consist of a cellular morphology as has been reported by Livingstone & Martin (3) and also by Mishra and Thomas (4). Alloys containing 1.5 w/o for 1 hour show a microstructure with 2:17 phase (~300Å in diameter) surrounded by 1:5 phase (~50 Å thick) in cellular configuration. The two phases are coherent and a typical image is shown in figure 5. This microstructure coarsens very slowly with aging for times of up to 10 hours or more. For aging times of ~ 30 hours, the 1:5 boundary phase is coarse (~500Å wide) and semicoherent and the 2:17 phase is larger in size (~ 3000 Å) as in fig. 6. Thus the increase in iH_c for isothermally aged alloys in fig. 4 is due to the fact that the 1:5 phase becomes a more effective barrier to the domain wall motion for thicker (but continuous) cell walls.

Step aging of the 1.5 wt/o Zr alloys must affect the chemical composition difference between the 1:5 and 2:17 phases, since the morphologies of the

isothermally aged the corresponding step aged alloys are indistinguishable. Clearly, the optimum morphology in this alloy is obtained after aging the alloy for ~ 15 hours isothermally at 850°C and the subsequent step aging changes the chemical composition so as to increase the iHc from 5kOe to ~ 15 kOe in fig. 4.

In 3% Zr alloys, the microstructures are again very similar. In the isothermally aged alloys, the scale of the microstructure in the 3% Zr alloys is finer than that of the 1.5% Zr alloys. The rate of coarsening for aging times of ~ 30 hours is only slight, unlike the 1.5 w/o Zr alloy case. The 2:17 cell are $\sim 600-1000\text{\AA}$ in size and the cell walls are $\sim 100\text{\AA}$ in width for alloys aged for ~ 30 hours (fig. 7). The changes in iHc due to step aging can be explained by assuming that the changes in the chemical composition plays the decisive role. The morphology for the 3% Zr alloys, aged for ~ 3 hours appears to be the optimum morphology.

Initial EDX data from STEM shows that the Cu:Fe ratio in the 2:17 and 1:5 phases change with step aging, leading to an enrichment of Cu in the cell boundary and enrichment of Fe in the cell after step aging. The distribution of Zr between the two phases is still uncertain

DISCUSSION

Some general conclusions for materials design can be drawn from the above results. Step aging of the alloys containing $\sim 2\%$ Zr is most beneficial for improving iHc as well as iHc (fig. 2) Large amount of Cu tends to increase the iHc, but reduces the squareness of the demagnetisation curve and hence $(BH)_{max}$ (fig. 1), and 25% Sm is necessary for larger $(BH)_{max}$. 4% Cu, 20% Fe alloy shows adequate iHc (fig.3) and can have better squareness (fig. 1). Thus an optimum composition for best hard magnetic properties would be $\sim 2\%$ Zr, $\sim 25\%$ Sm, $\sim 20\%$ Fe and $\sim 4\%$ Cu. The demagnetisation curves for such an alloy is shown in Fig. 8, along with that for a 26% Sm, 6% Cu, 15% Fe, 2.5%Zr alloy and the best magnetic properties, viz: $(BH)_{max} = 33\text{MGoe}$, $iHc = 13\text{kOe}$, $Br = 12\text{kG}$, are obtained for the 4% Cu, 20%Fe alloy, after step aging. Such a magnet combines the optimum chemical composition for partitioning of various elements between the 1:5 and 2:17 phases, and also the optimum cell diameter to boundary phase thickness. Further insight to improve the $(BH)_{max}$ may be obtained by the study of the influence of the chemical composition and processing conditions on the microstructure in greater detail.

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