MAGNETIC PROPERTIES OF Fe-6.4wt%Si RIBBONS

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ABSTRACT - Thin Fe-6.4wt.%Si ribbons were produced by melt spinning. High temperature recrystallizations, performed at 1025°C in a hydrogen atmosphere, were found to produce the lowest H_c values (19 A/m). Further agings were carried out at 50°C intervals in the range 400-700°C to optimize the magnetic properties. For all ribbons we measured H_c (60 Hz and DC), the maximum permeability μ_{max}, the saturation magnetostriction λ_s, and the effective anisotropy constant K_{eff}. In general, the agings did little to improve the magnetic properties, and those around 600°C resulted in their deterioration. Extensive TEM investigations of the ribbons indicate that the dendritic structure of the as-cast material disappears after recrystallization, leading to a more uniform distribution of Si as well as a more homogeneous ordering. The 600°C aging results in a marked anisotropy in the B2 antiphase boundaries and the growth of oxide particles, which lead to a deterioration of the magnetic properties.

I. INTRODUCTION

Adding silicon to electrical steels has become common because silicon increases the electrical resistivity with a consequent reduction in magnetic losses. For compositions around 6.5wt%Si, the magnetostriction is practically zero and hysteresis loss is minimum. However, for compositions above 4.5wt%Si there is a drastic reduction in the ductility of the alloy, making it impossible to use conventional casting and cold rolling techniques. The development of rapid solidification technology, on the other hand, has made it possible to obtain ribbons of Fe-6.5wt%Si with excellent magnetic properties [1].

For compositions above about 5wt%Si, the disordered bcc structure (A2) gives rise to ordered phases with the CsCl (B2) or the $Fe_3Al(DO_3)$ structures [2]. These phases in principle influence the mechanical and magnetic [3] properties of the ribbons and their presence has been commented on by various authors. *Narita et al.* [4] studied the effect of the order-disorder reaction on sheets of Fe-6.5wt%Si which had been forged and rolled, concluding that an aging at 500°C was more effective in improving the magnetic properties than treatments at higher temperatures, which presumably produced B2 ordering. X-ray diffraction spectra indicated DO_3 order caused by the aging at 500°C. More recently, *Degauque et al.* [3] studied the influence of ordering on the coercivity and hysteresis loss of rapidly solidified Fe-6.5wt%Si. These authors found losses to be lower in ribbons aged at 700°C than in those aged at 500°C. TEM showed that the high temperature aging (700°C) promoted the growth of the B2 structure, which was beneficial for the magnetic properties. In the present work, we study the effect of aging treatments in the temperature range 400-700°C on the microstructure and magnetic properties of Fe-6.4wt%Si ribbons produced by meltspinning. Extensive TEM investigations were carried out to follow the microstructure changes induced by these treatments.

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II. EXPERIMENT

Master ingots were prepared by arc melting electrolytic iron (99.99%) and pure silicon (99.999%) under an argon atmosphere. Ribbons were produced by planar flow casting onto a low carbon steel wheel, using flowing argon gas to protect the ribbons from oxidation. Continuous ribbons of 3mm width and 20μm thickness were produced with exceptionally clean surfaces, indicating the absence of oxidation. Wet chemical analysis of the ribbons indicated 6.4wt%Si, 0.022%C, 0.006%Mn, 0.003%Al, and 0.003%P. The as-cast ribbons were cut into 150mm long pieces and recrystallized between 950 and 1150°C for 1h in flowing H_2 gas (1 mbar). Ribbons recrystallized at 1025°C for 1h were then annealed for 2h between 400 and 700°C, also in a H_2 atmosphere, in order to develop the B2 and/or DO_3 ordered structures.

Measurements of the coercive field (60 Hz and DC) and the maximum permeability (μ_{max}) were made with a hysteresis loop tracer on 150mm long samples inserted into a solenoid. The saturation magnetization (M_s) was obtained from a vibrating sample magnetometer (VSM) and the saturation magnetostriction λ_s was determined using the small-angle-magnetization-rotation method [5]. The effective anisotropy energy was obtained from the initial magnetization curve using the relation

$$K_{eff} = \int (M_s - M)dH \quad (1)$$

Microstructural characterization was carried out with a JEOL TEMSCAN-200kV having EDAX capability. The samples were prepared by mechanical polishing, followed by thinning in an electrolyte consisting of 5% perchloric acid in 2-butanoethanol (T = -10°C, 50V).

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III. RESULTS AND DISCUSSION

TEM observations made on as-cast ribbons showed high dislocation density and significant inhomogeneity in the Si content along the ribbon (EDAX analysis indicated (6.1 ± 2.2)%Si.) associated with the dendritic structure, which, in turn, results in an uneven distribution of the B2 antiphase domains (Figure 1a). The average grain size in as-cast material was 10μm. Small (≈0.1μm) domains with B2 ordering were observed, but DO_3 reflections were very weak. The magnetic properties of as-cast ribbons were: M_s = 1.85T, T_c = 700°C, λ_s = 2.9×10^{-7}, H_c(60Hz) = 100A/m, H_c(DC) = 95A/m, and μ_{max} = 4000.

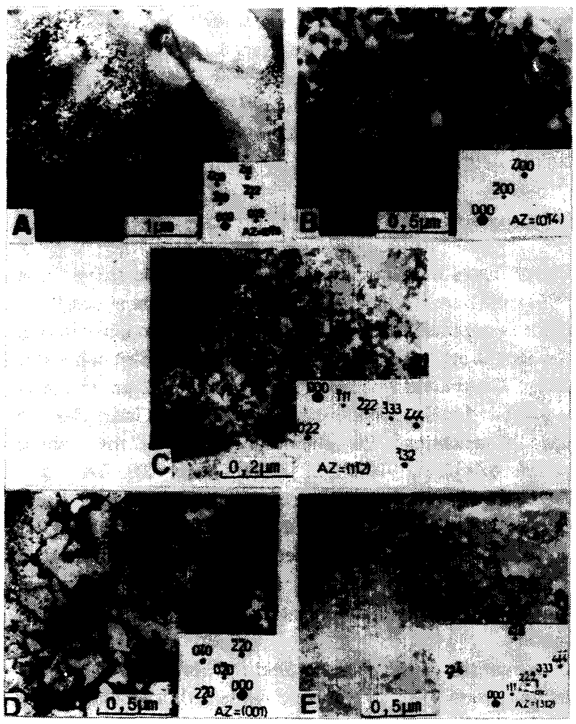

Figure 1 - Superlattice reflections and dark fields: a) As cast, B2 order; g = $\bar{2}00$; b) After recrystallization, B2 order; g = $\bar{2}00$; c) After 2 hours at 500°C, DO_3 order; g = $\bar{1}11$; d) After 2 hours at 600°C, B2 order; g = $0\bar{2}0$; e) After 2 hours at 600°C, oxides.

Figure 2 shows the coercivity as a function of recrystallization temperature. For 1h treatments in a H_2 atmosphere of 1 mbar, the lowest coercivity values were obtained at 1025°C: H_c(60Hz)=37A/m, H_c(DC)=19A/m, and μ_{max} = 16500. TEM observations showed a more uniform distribution of Si as a consequence of the disappearance of the dendritic structure in the recrystallized material. The dislocation density also decreased considerably. The average grain size increased to 170μm and the B2 regions became larger (≈0.3μm). As can be seen in Figure 1b, the distribution of B2 antiphase boundaries is isotropic. Reflexions associated with DO_3 ordering were very weak. X-ray diffraction spectra of as-cast and recrystallized ribbons indicated the development of (001)[0kl] texture during the heat treatment at 1025°C. The significant improvement of the magnetic properties is associated with the increase in grain size and the development of texture, since both act in the direction of increasing the domain wall mobility.

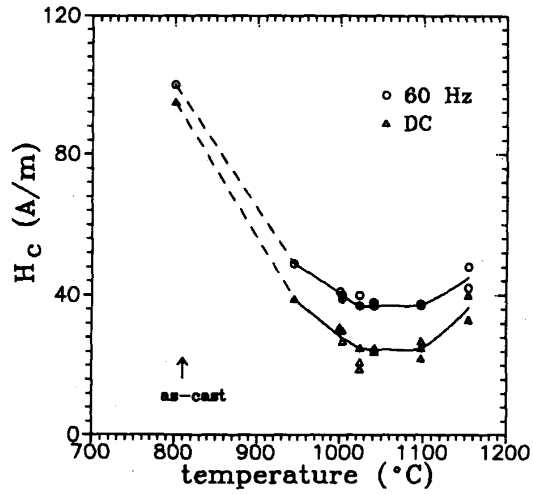

Figure 2 - Coercive field H_c vs recrystallization temperature.

After the recrystallization at 1025°C, a series of aging treatments was carried out to examine the ordering process and its effect on the magnetic properties. Figures 3 and 4 show the effect of aging on the induced anisotropy constant K_{ind}, the maximum permeability μ_{max}, the coercive field H_c, and the magnetostriction λ_s. In general, the aging treatments cause little change in the magnetic properties, except for temperatures around 550-600°C. The effective anisotropy results from several contributions, including the magnetocrystaline anisotropy (K_1) and a magnetoelastic term ($(3/2\lambda_s\sigma)$). Since $\mu \approx 1/K_{eff}$ and $\mu \approx 1/H_c$, we expect an increase in K_{eff} to correspond to an increase in the coercivity and a decrease in the permeability. In fact, our results show that an increase in K_{eff}, ocurring for temperatures above 550°C, is accompanied by a decrease in μ_{max} and a substantial increase in H_c. As can be seen in Figure 4, λ_s is small, except for aging temperatures around 550°C and 600°C where it increases to values about 10 times larger.

TEM observations of ribbons aged at 500°C show a stronger presence of DO_3 ordering. (See Figure 1c.) The B2 domains are present and of the same size as in the recrystallized ribbon. The grain size has not been altered by this aging, but we note the presence of some very small oxide particles, which are uniformly distributed.

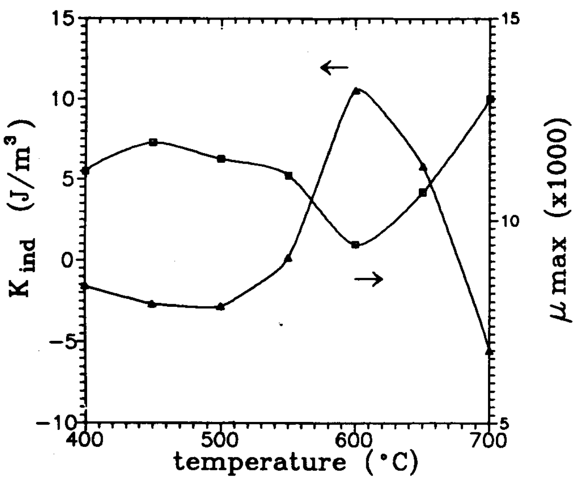

Figure 3 - Induced anisotropy K_{ind} and maximum permeability μ_{max} vs. aging temperature.

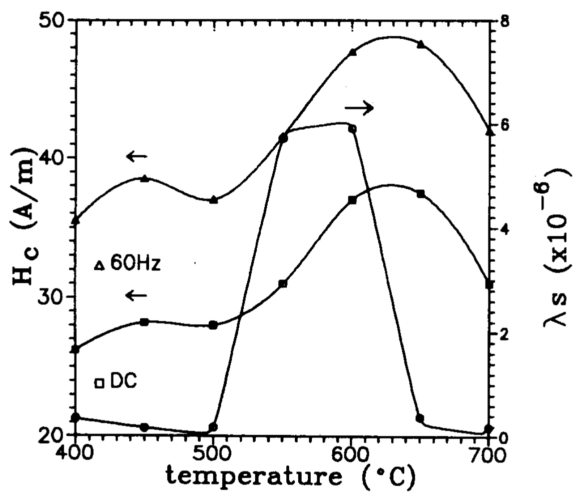

Figure 4 - Coercive field H_c and magnetostriction λ_s vs. aging temperature.

For the ribbons aged at 600°C, substantial changes are observed in the microstructure. The regions corresponding to B2 ordering have increased in size to about 0.5μm. More striking is the fact that the 1/4[111] antiphase boundaries of B2 show a marked anisotropy, having a preference for the [100] planes. (See Figure 1d.) Furthermore, we notice the presence of DO_3 ordering and a reduction in the number of oxide particles. These, however, increase in diameter (≈0.03μm), attaining a size which is comparable to the thickness of a magnetic domain wall. (See Figure 1e.) The overall grain size remains the same, however. Ribbons aged at 700°C showed B2 antiphase boundaries, which are again more isotropic, as well as a smaller number of larger oxide particles.

The deterioration of the magnetic properties in our ribbons for the 600°C aging is seen to be simultaneously accompanied by a marked anisotropy in the 1/4[111] B2 antiphase boundaries and by the presence of oxide particles of size comparable to the magnetic domain wall thickness. The authors of Ref. 2 observed a strong anisotropy in the 1/2[100] DO_3 domain boundary morphology, which disappeared as the temperature increased. For our ribbons, we observe an anisotropy at a temperature near the boundary between the B2 and B2 + DO_3 phase fields, according to the phase diagram of Ref. 2. Strong local fluctuations in the degree of long range order at the A2 - B2 and B2 - DO_3 boundaries have been observed in Ref. 2. In our case, similar fluctuations may have promoted the development of the observed anisotropy. In summary, we have seen that the aging treatment at 600°C results in the presence of induced anisotropy and oxide particles with dimensions comparable to a domain wall thickness, which, leading to a reduced mobility for the magnetic domain walls, has a negative effect on the magnetic properties.

IV. CONCLUSIONS

Melt-spun ribbons of composition Fe-6.4wt%Si were recrystallized at 1025°C in a hydrogen atmosphere and were then subjected to aging treatments in the temperature range 400-700°C. Extensive TEM investigations indicate that the dendritic microstructure of the as-cast material disappears after recrystallization, leading to a more uniform distribution of Si as well as a more homogeneous ordering. A marked anisotropy in the B2 antiphase boundaries is accompanied by a deterioration of the magnetic properties after a 600°C aging. The presence of a large number of oxide particles also contributes to decreasing the domain wall mobility.

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