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Evolution of magnetomechanical damping and magnetic properties of pure iron siliconized up to 5.5 wt.% by chemical vapour deposition

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Abstract

With a high silicon content, silicon steel has good soft magnetic properties because of the decreased magnetic anisotropy, lower magnetostriction and higher resistivity. However, above 3.5 wt.% Si a lower ductility makes conventional processing into textured strips extremely difficult. In this investigation we consider another possible method which consists of depositing silicon and aluminium on a pure iron substrate by chemical vapour deposition. The magnetomechanical damping evolution (measured at 0.7 Hz with a torsional pendulum) of enriched iron strips containing between 0.5 and 5.5 wt.% Si + Al is considered. Results show a strong decrease in magnetomechanical damping with increasing Si + Al content, which coexists with a lowering of the magnetic core losses measured at 50 and 400 Hz. Results relative to as-prepared and homogenized states are compared.

1. Introduction

Silicon steel is particularly suitable for transformers and motor cores because of its good soft magnetic properties. With 6.5 wt.% Si content this material has improved performance because of the higher resistivity, lower magnetostriction and decreased magnetic anisotropy resulting from the increased silicon content. However, above 3.5 wt.% Si a lower ductility makes conventional processing into textured strips extremely difficult. Chemical vapour deposition (CVD) is one convenient method which is capable of depositing a large variety of metals, alloys and metallic compounds on various substrates. Thus this method has been proposed to deposit silicon and/or aluminium on an iron substrate in order to improve the magnetic properties of iron [1–3]. The addition of Si and/or Al to the substrate surface changes the composition locally and improves the alloy properties [4, 5]. In this paper, after a short description of the CVD process used for pure iron siliconization, we examine the magnetomechanical damping evolution of iron strips which have been enriched with up to 5.5 wt.% Si + Al.

2. Elaboration and experimental processes

The CVD process (pack cementation) used in this study consists of standing the samples to be coated upright in a semisealed retort filled with an appropriate mixture of iron silicide and aluminide as source (15 wt.% Si, 4 wt.% Al, with minor amounts of Ti, Cr, Cl and P) and a halide activator salt such as NH_4F. Slow heating to 850 °C enables the activator to sublimate and to decompose as

$$NH_4F(g) \longrightarrow NH_3(g) + HF(g)$$

Hydrogen fluoride reacts with the source in the retort as

$$2HF(g) + Si/Al(s) \longrightarrow Si/Al \text{ fluorides} + H_2(g)$$

The siliconizing and aluminizing atmosphere is then ready for operation and the deposition process begins. The metal halide molecules diffuse through the pack to the substrate, where surface reactions deposit the metallic components for diffusion into the substrate [6, 7]. Pure iron strips (size $100 \times 10 \times 0.3$ mm^3) are chemically polished in an oxalic hydrogen peroxide aqueous solution and rinsed with deionized water; then they are introduced with the iron silicide powder into a

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cylindrical retort containing the halide activator salt. Codeposition treatment is performed at the same time at 1000 °C in a hydrogen atmosphere. After the deposition the retort is cooled rapidly to room temperature under hydrogen, which is used as a protective gas in order to avoid oxidation effects. The characteristics of the coatings are determined by weighing, optical microscopy and electronic microprobe spectroscopy.

Magnetomechanical damping measurements are performed at room temperature at 0.7 Hz frequency with an inverted torsional pendulum. The damping is measured as a function of the shear strain γ, which varies between 1×10^{-5} and 24×10^{-5} at the surface of the sample. Then the values of γ are corrected in order to take into account the heterogeneity of the torsional deformation [8]. This correction induces a slight increase in the maximum amplitude and a slight shift towards small deformation, but it does not change the relative shapes and amplitudes of the initial curves.

The magnetomechanical damping $Q_{mag}{}^{-1} = Q^{-1}(H=0) - Q^{-1}(H_s)$ is defined as the difference between the damping in the demagnetized state and the damping under the saturating field ($H_s = 80$ kA m^{-1}). The internal friction background is always about 5% smaller than the magnetomechanical damping. Before each measurement the sample is demagnetized.

3. Experimental results and discussion

The CVD leads mainly to the formation of a non-porous, uniform and adherent iron–silicon–aluminium solid solution (Fig. 1). The composition of the solid solution depends on the experimental conditions.

Figure 2 plots the variation in the magnetomechanical damping $Q_{mag}{}^{-1}(\gamma)$ in the as-prepared state for five Si + Al contents determined by weighing assuming a uniform distribution. The damping maximum quickly

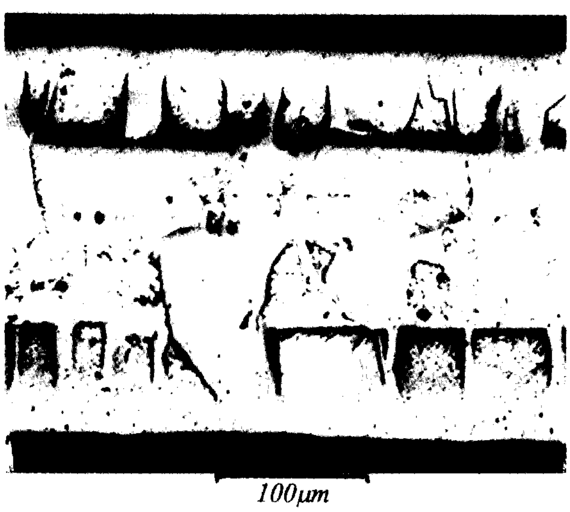

Fig. 1. Cross-section of iron sample treated at 1000 °C with NH_4F for 2 h.

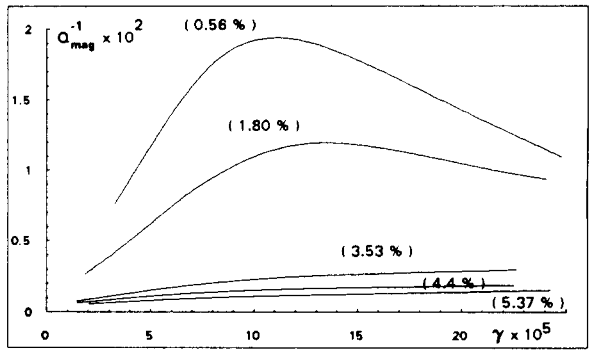

Fig. 2. Magnetomechanical damping maximum variation $Q_{mag}{}^{-1}(\gamma)$ of iron strips with various values of Si + Al content in as-prepared state.

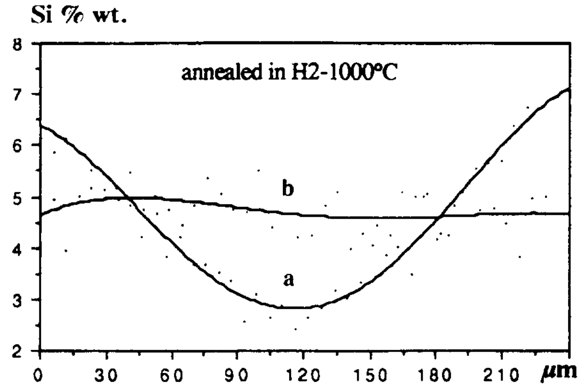

Fig. 3. Evolution of Si concentration gradient during diffusion annealing of pure iron pretreated at 1000 °C for 4 h in NH_4F environment: a, as-prepared state; b, homogenized state after 10 h at 1000 °C under H_2 flow.

decreases then vanishes when the Si + Al content increases above 5.5%. In fact, in the as-prepared state, because of the concentration gradient (Fig. 3), Si and Al are mainly located at the outer surfaces of the iron strips.

Figure 4, corresponding to the three higher contents, gives the evolution of the maximum damping after two durations (2 and 10 h) of diffusion annealing at 1000 °C under an H_2 flow. In comparison with the as-prepared state, for 4.4 and 5.37 wt.% Si + Al full homogenization induces an increase in magnetomechanical damping. In particular, for 4.4 wt.% Si + Al the observed increase is a maximum and reaches more than 500%.

Figure 5 shows a comparison of the damping maximum $Q_{mag}{}^{-1}(\gamma)$ between the three homogenized states and pure iron as well as a commercial Fe–3 wt.% Si alloy (thickness 0.3 mm) with randomly oriented grains annealed in an H_2 environment.

For the iron strip the large grain size (1 mm) and high purity promote the mobility of the 90° magnetic domain walls and thus promote substantial magnetomechanical hysteresis losses and large maximum damping [9]. In spite of a smaller grain size and lesser purity, the commercial FeSi alloy shows significant damping

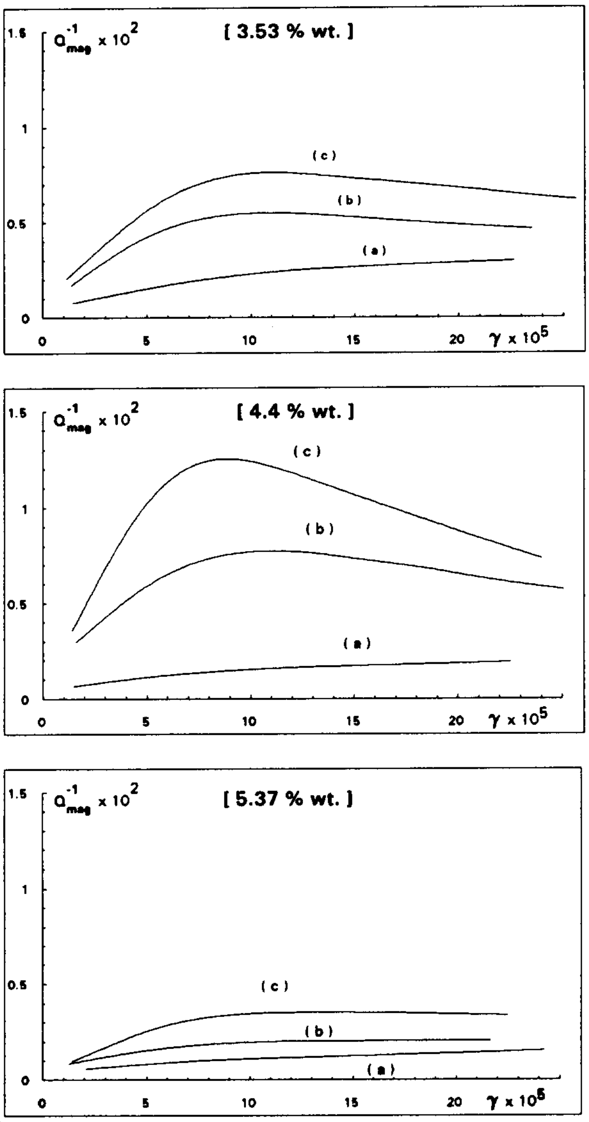

Fig. 4. Evolution of $Q_{mag}{}^{-1}(\gamma)$ for various Si + Al contents with diffusion annealing at 1000 °C under H_2 for (a) 0 h, (b) 2 h and (c) 10 h.

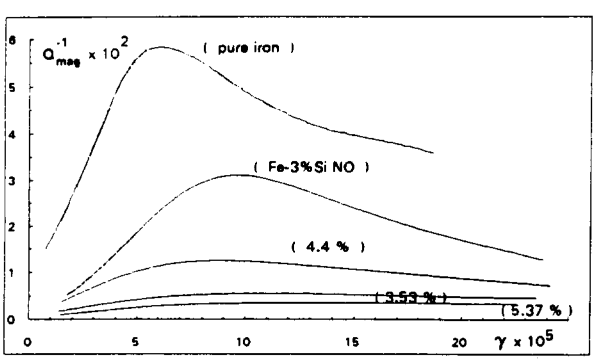

Fig. 5. Variation in magnetomechanical damping maximum $Q^{-1}(\gamma)$ of 3.53, 4.4 and 5.37 wt.% Si + Al iron strips after homogenization (10 h at 1000 °C in H^2) compared with pure iron and non-oriented Fe–3 wt.% Si.

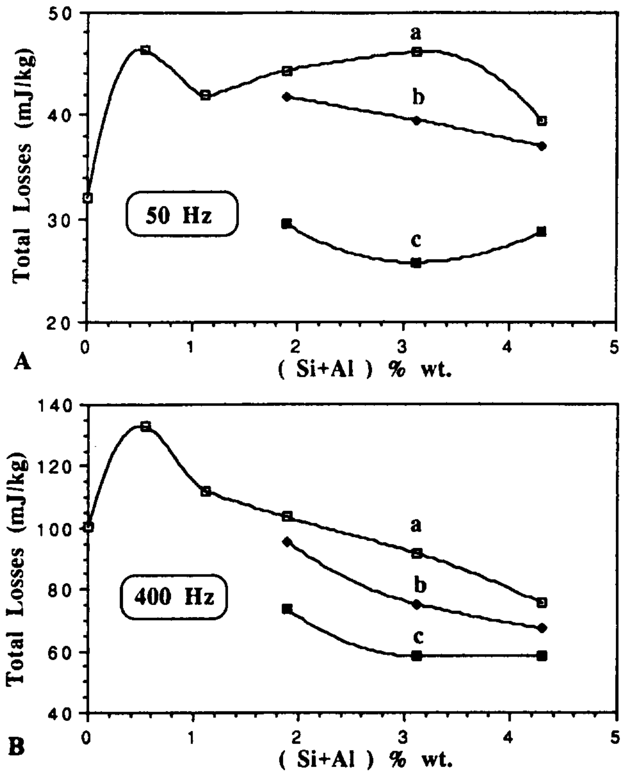

Fig. 6. Evolution of magnetic losses as a function of Si + Al content measured at 1 T induction and medium frequencies of (A) 50 Hz and (B) 400 Hz after diffusion annealing at 1000 °C under H_2 for (a) 0 h, (b) 2 h and (c) 10 h.

because of the magnetostriction value of iron, which reaches its maximum at about 3 wt.% Si [10]. Owing to the composition of the mixture source used for the coating treatment, the CVD strips contain approximately 2.5 times as much Si as Al. Consequently, the 3.5, 4.4 and 5.37 wt.% Si + Al homogenized samples contain about 2.5, 3.1 and 3.8 wt.% Si respectively, leading to the relative ordering of the magnetomechanical curves in Fig. 5, which shows a higher damping for the 4.4 wt.% Si + Al content. Aluminium also increases the magnetostriction [10]; however, for our three samples the aluminium content remains too weak to exert a significant influence on the relative ordering, which is controlled by the silicon content. Lastly, in comparison with the commercial Fe–3 wt.% Si alloy, the weak magnetomechanical damping level for the 3.53 and 4.4 wt.% Si + Al samples suggests that the CVD treatment creates structural alterations opposing the stress-induced and irreversible displacement of the non-180° domain walls. This decrease in the mobility of the 90° domain walls promotes weak magnetomechanical hysteresis losses.

Other sources of magnetic losses are eddy current power losses which appear when an a.c. magnetic field is applied to magnetize a ferromagnetic body. Their occurrence gives rise to energy losses W_e which are directly proportional to the square of the frequency of the driving magnetic field and inversely proportional to the electrical resistivity of the material. Both silicon and aluminium increase the resistivity and consequently decrease the eddy current losses (Figs. 6A and 6B) of iron at low frequency (50 Hz) and medium frequency (400 Hz) measured at 1 T. It is seen that complete homogenization of the silicon and aluminium distribution within the iron strips promotes lower magnetic losses, thus making enriched silicon steel particularly suitable for the production of transformers and motor cores.

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