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Research articles

Surface modification and its role in the preparation of FeSi gradient alloys with good magnetic property and ductility

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ABSTRACT

Realization of the effective Si penetration at a lower processing temperature is a challenge, but of significance in reducing the strict requirements for the equipment and realizing cost-cutting in production. In this work, we have modified the surface microstructure of Fe-3 wt%Si alloy by using surface mechanical attrition treatment. The modified surface microstructure is characteristic of nanocrystalline, which is found to significantly enhance the efficiency of subsequent Si penetration into the alloy, and successively leading to the decrease of penetration temperature up to 200 °C. As a consequence, the Si gradient distribution across thickness can be readily controlled by changing penetration time, and FeSi alloys with various gradients are prepared by chemical vapor deposition along with subsequent annealing process. The dependence of magnetic and mechanical properties on Si gradient for demonstrates that the increase of Si gradient reduces core losses, especially at higher frequencies, and meanwhile improves ductility of FeSi alloys as well. The mechanism underlying the effect of Si gradient is clarified by combining magnetostriction measurement and domain structure observations. This work provides a facile and effective way for achieving gradient FeSi alloys with good magnetic property and ductility.

1. Introduction

FeSi alloys with 6.5 wt% Si possessing excellent magnetic properties are much attractive as core materials in the applications of transformers, high speed motors and electric power [1,2]. The high Si content in the alloy is often achieved by the penetration of Si into FeSi alloys with lower Si content [3–6]. Among existing penetration methods, chemical vapor deposition (CVD) along with subsequent Si diffusion process has proved most advantageous from the standpoint of commercial scale production and non-limitation on sample shape [6]. However, during the CVD process, high temperature of ≥1100 °C is required to guarantee the reaction between $SiCl_4$ and Fe as well as the subsequent Si diffusion to be performed at a certain rate [7], which puts forward strict requirements for the equipment, and leading to high cost of production as well as production difficulty. Realization of effective Si penetration at a lower processing temperature is of scientific interest and practical significance for overcoming the problem, which is, however, very challengeable due to the very slow diffusion of Si below 1100 °C.

It is well known that grain boundary diffusion possesses a larger rate than that of intragrain. The decrease of grain size can provide more of the diffusion channels caused by the higher density of grain boundary. Meanwhile, the higher energy arising from more boundaries is in-stored in the system, which is also expected to be favorable for improving diffusion rates [8–10]. In this respect, we have proposed to refine the grains on Fe-3Si% sample surface by applying surface mechanical attrition treatment (SMAT) [11], in an attempt to increase the deposition and diffusion rates at lower temperatures. On the other hand, the gradient FeSi alloys, characteristic of gradual reduction in Si content from surface to interior, have attracted much attention for their better workability while containing high average Si content, as compared to the conventional counterparts with homogeneous Si distribution [12–15].

In this work, we have applied the SMAT approach in an attempt to lower the temperature of the Si penetration performed by using the chemical vapor deposition (CVD). Fe-3 wt% Si starting sheets were applied to prepare the FeSi alloys with different Si gradient profile across thickness. The Si penetration temperature has been effectively decreased by up to 200 °C for the SMATed alloy as compared to the unSMATed. Effect of Si distribution on the related properties is studied in an attempt to realize simultaneous improvement in the magnetic property and ductility of FeSi alloys.

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Mechanism underlying the role of Si gradient is discussed by combining magnetostriction measurement and domain observations.

2. Experimental

SMAT technique was applied to refine surface microstructure of commercial grain-oriented Fe-3Si (wt%) (35Z155) with the initial grain size of 3 mm on average. The size of the starting alloy is $0.35 \times 8 \times 40\ \mathrm{mm}^3$. Prior to Si deposition, Fe-3Si alloys were subjected to the SMAT process in a ball-milling chamber. Stainless steel balls of 5 mm Ø AISI 316L were used for collisions onto the sample surface, and the balling process was performed at a fixed frequency of 20 Hz. The penetration of Si into the samples was performed by CVD method along with subsequent diffusion processes. Firstly, Fe-3Si alloys were heated to the deposition temperatures ranging from 800 to 1180 °C in a highly pure N_2 atmosphere; secondly, a gaseous mixture of $SiCl_4/N_2$ was introduced into the chamber in which the reactions between the vapor reactants and samples are expected to take place. The deposition time was set to be 10 min. After the deposition, the samples were subjected to diffusion processes without being removed out of the chamber at the same temperatures for various time in the range of 30–120 min.

Surface and cross-sectional microstructures for the SMATed alloys were observed using transmission electron microscopy (TEM) and electron backscattered diffraction system (EBSD), respectively. The EBSD measurement was carried out at an accelerating voltage of 20 kV and a step size of 15 nm. Si distribution across thickness was measured by electron dispersive spectrometer (Link ISIS).

Automatic B-H curve tracer was applied to evaluate hysteresis loss, while alternating current core loss analyzer was used to measure total core loss with a controlled sinusoidal induction under a magnetic field of 0.5 T in the frequency range of 1–10 kHz. Finite element method (FEM) was used to simulate distributions of eddy currents across thickness of the alloys and to calculate classical eddy current losses. To make the simulation more reasonable, variations of resistivity and permeability should be considered as a function of Si distribution. Magnetic force microscopy (MFM) was applied to observe magnetic domain structure with Icon-Veeco atomic force microscope, and magnetostriction was measured by standard strain gauge method. Micro-hardness was measured on FM-800 tester under a load of 200 gf for 15 s. Elongation were measured to evaluate ductility performance using Instron 5565 tester at a strain rate of 0.5 mm/min. The active length of the sample for the measurement was 20 mm, and the size of the part bitten by the jaws was about 8 mm.

3. Results and discussion

3.1. Effect of SMAT on Si penetration

Fig. 1(a) shows cross-sectional EBSD image across the area of 0–11 μm from surface for the SMATed Fe-3Si sample, showing gradient microstructure along the cross-section. The grain size is nanoscaled in the area across 0–1μm from surface (marked by arrow). The surface grain size can be estimated to be smaller than 100 nm, from the TEM image in Fig. 1(b). While in the region of 1–4 μm from the surface, the microstructure is characteristic of larger and elongated grains. Furthermore, in the region deeper than 4 μm from the surface, microstructure is shown to be the same as that of the unSMAT sample. The grain size distribution is clearly attributed to the various extents of plastic deformation across thickness caused by the SMAT process [16,17]. The larger deformation at surface area is responsible for the formation of nano-scaled and equiaxed grains. Fig. 1(c) demonstrates the variation of Vickers hardness across the thickness, showing a maximal value of 340 on the surface, followed by a dramatic reduction to the hardness of 210 at 80 μm from the surface. The hardness is 250 at 11 μm from surface, still larger than that of the unSMATed, which indicates that the defects caused by deformation also exist in the area even without grain size variation.

Fig. 2 displays effect of surface nanocrystallization on the Si cross-sectional distribution near surface area for the Si-deposited samples prior to annealing processes. For the unSMATed sample deposited at 980 °C, only a slight of Si is detected in the region of 3 μm from the surface. While for the SMATed alloys, it can be seen that Si is effectively penetrated into the deeper zone. Moreover, the Si penetration is increased with the longer SMAT processes. When the treatment time increases to 120 min, the Si distribution for the 980 °C deposited sample (curve D) is about the same as that of the unSMATed one deposited up to 1180 °C (curve E). Our preliminary work has shown that the increased SMAT time results in a stronger refinement of grains near the surface area. The great improvement in the efficiency of Si deposition due to the SMAT is obviously associated with the grain refinement which brings about more grain boundaries. In order to highlight the role of surface nanocrystallization, we defines the penetration efficiency as $\eta = \frac{C_{processed} - C_{origin}}{C_{origin}} \times 100\%$, where C_{origin} and $C_{processed}$ are the surface Si contents before and after Si penetration for the samples, respectively. As listed in the inserted table, the penetration efficiency of the SMATed for 120 min is 117% at 980 °C, very close to 120% for the unSMATed when deposited at 1180 °C. It indicates that the SMAT approach allows for a great reduction of the deposition temperature for a given penetration efficiency.

In order to optimize the deposition temperature, the change of Si distribution with the temperature for the samples prior to annealing process is shown in Fig. 3(a). The Si penetration is monotonously increased with increasing the temperature from 800 to 980 °C. As the temperature further increases to 1080 °C, however, a reduction of the penetration is seen. Fig. 3(b) and (c) show the EBSD images for the SMATed alloys deposited at 980 and 1080 °C, respectively. It is clearly seen that the depositions at both temperatures lead to grain growth. Nevertheless, the grain size of the samples deposited at 980 °C is still less than 1 μm, while those deposited at 1080 °C show the grains with the similar size to that of unSMATed alloys. The large grain growth decreases grain boundaries, which is responsible for the reduction of penetration. The result supports the argument that grain boundaries play a key role in the Si penetration for SMATed alloys.

The as-deposited alloys were then subjected to annealing process at 980 °C for different times in order to optimize the Si gradient distribution. As shown in Fig. 4, the annealing time has a great influence on the Si penetration. When the annealing time is 30 min, only slight penetration of Si is detected around the sample surface area. As the annealing time increases to 60 min or longer, the Si is shown to diffuse into the interior of alloy and the gradient Si distribution is achieved. The gradient decreases with increasing the annealing time, showing a maximal value and the surface Si content of 6.5 wt% at 60 min. As the time increases to 120 min, the Si distribution becomes nearly homogeneous. It is further revealed that the distribution can be fitted with the parabolic formula $C(x) = Ax^2 + B$, where x is the distance from the center. Based on $C'' = \frac{\partial^2 C(x)}{\partial x^2}$, the Si gradient ($C''$) is estimated to be 2.1×10^{-4} and 0.3×10^{-4} when annealed for 60 and 120 min, respectively.

3.2. Magnetic properties of the gradient alloys

Fig. 5(a) displays core losses for the samples with various Si gradients as a function of frequency, under the magnetic induction of