Diffusing Behaviors of Si in Silicon Steel during Annealing Process

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Abstract

This paper studies on the influence of annealing temperature on the distribution of Si content in the electro-deposition samples of high silicon steel, and the content distribution of Si, the section layer textures and the texture distribution are also analyzed. The results show that higher the temperature is, bigger the diffusion coefficient gets. When the temperature reaches 1050 C, the annealing time is 210min, in which case the crystals in the samples are distributed well. Si is also well distributed, with the average concentration 6.3715%. Analyzing the textures in the samples, the content of easily magnetic textures of {100} and {110} with high density increases, while the density of bad textures decreases.

Keywords: *silicon steel, diffusion coefficient, annealing, texture*

With the increase of silicon content, the increased brittleness of silicon steel makes the subsequent processing of materials difficult. It has found that 6.5wt%Si steel is the ideal material for the iron core in high frequency, low noise, and low core loss [1]. In order to obtain high silicon steel sheet which can be a good magnetic properties of silicon, researches for new technology have been become a pressing matter at the moment. The molten salt electro deposition for making high silicon steel can avoid the problems of rolling difficulties. The test used cold-rolled silicon steel sheet as the base to get high silicon content by electro-deposition in molten salts. By high temperature diffusion annealing obtains the high silicon steel sheet that Si concentration distribution. Research results [2] show that: The diffusion activation energy and diffusion frequency factor of Si in Fe-Si are associated with not only the temperature but the Si content in the alloy. Derived by computer simulations that the effect of temperature on the diffusion coefficient is great[3]. At 1100 C the diffusion coefficient is two orders of magnitude bigger than the high diffusion coefficient at 900 C. The analysis results of Mehrer[4] are similar with the above research results. Fe, Si atoms diffusion and transfer ability changed with the environment and temperature in Fe-Si thin films [5]. In current study, the concentration of Si changed with the thickness in the annealed samples, and the relationship between temperature and Si atomic diffusion coefficient is linear through the theoretical calculation. To determine the optimal annealing temperature this study analyzes the microstructure and texture of the annealing sample under different temperatures, to form the theoretical basis for the preparation of silicon steel sheet with high silicon content.

1 Experiment Methods

The material in the experiment is non-oriented silicon steel sheet with 1.6% silicon consent, $20\times20\times0.5mm^3$. Measure the experiment temperature with platinum rhodium-platinum thermocouple, with the error range of ±10 C. Make the silicon steel sheet as cathode. Put the graphite with $NaCl\text{-}KCl\text{-}NaF\text{-}SiO_2$ into the resistance furnace to heat. The electro-deposition time is 60min and $50mA\cdot cm^{-2}$ for the current density. The front-to-back ratio of pulse in current study is 9:1 and the electro-deposition temperature is 800 C. Take the deposited samples into the 900～1050 C SRAF to diffuse and anneal. The GDA-750 GDS, JXP-10 metallographic microscope and D5000 X-RAR DIFFRACTOMETER were used to analyze and test these samples.

2 The Result and Analysis

2.1 Effect of annealing temperature on diffusion of Si content in samples.

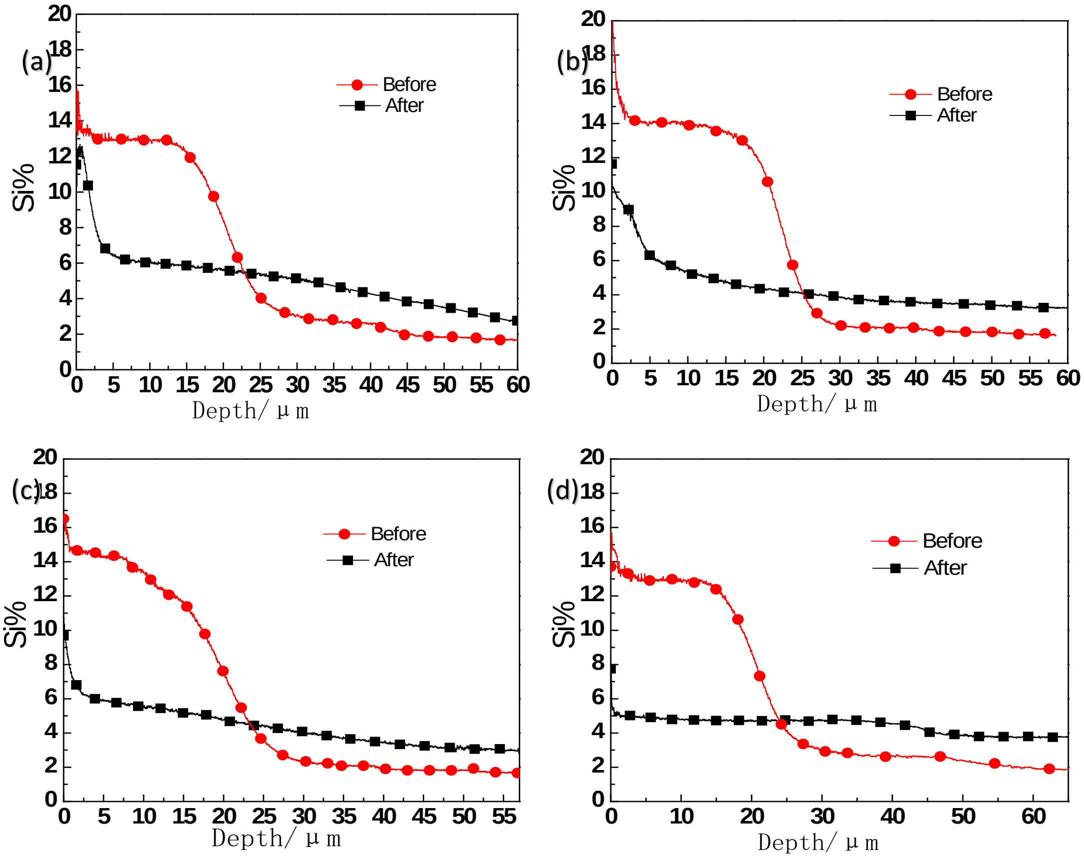

Figure 1 Effect of annealing temperature on distribution of Si content in samples (a) 900℃ (b) 950℃(c) 1000℃ (d) 1050℃

The concentration of Si content influenced by annealing temperature is shown in Fig 1. It is clear that the concentration of Si content changes greatly before and after annealing. High temperature annealing promotes the internal diffusion of Si in deposition layer. The Si content in surface layer decreases, while the depth of diffusion increases. When the temperature gets to 1050 C, the concentration tends to be more evenly distributed.

2.2 Diffusivity

In the process of annealing and homogenization diffusion, the samples after electro-deposition have obvious diffusing character: the total account of Si in samples is constant, and does not change with the change of diffusion conditions. As shown in the fig 1, the concentration of Si appears exponential decaying. With the increase of x, the concentration reduces rapidly. The shorter the diffusing time is, the higher the concentration gradient is. With the extension of diffusing time, the speed of reduction decreases, which means the formation of the concentration gradient decreases. So, it is clear that the diffusion process satisfies one dimensional film source unsteady diffusion model.

Take the concentration of Si content in electro-deposition as the result of that the components diffuse towards the substrate in the condition of annealing temperature for time t_0, the concentration distribution of the samples after annealing as the concentration distribution of that the components diffuse towards the substrate for time t_0+t. The changes of concentration are not higher than 20%, and the influence of the electro-deposition time on the diffusivity of Si has been proved. Use the Gaussian solution of Fick's second law to solve:

$$c(x,t) = A + \frac{M}{\sqrt{\pi D t}} \exp\left(-\frac{x^2}{4Dt}\right) \qquad (1)$$

The concentration distribution of Si content in the samples can be shown as:

$$c\left(x,t_0\right)=A+\frac{M}{\sqrt{\pi D t_0}}\exp\left(-\frac{x^2}{4Dt_0}\right) \qquad (2)$$

The concentration distribution of Si content in the samples after annealing can be shown as:

$$c\left(x,t_1\right)=A+\frac{M}{\sqrt{\pi D t_1}}\exp\left(-\frac{x^2}{4Dt_1}\right) \qquad (3)$$

In the above formulas,

c(x, t_0) ---- the concentration of Si content corresponded to the depth x after the electro-deposition %.

c(x, t_1) ---- the concentration of Si content corresponded to the depth x after annealing %.

M ----the total amount of diffusion components of Si content %.

A ---- constant. During the calculation, the concentration of Si content is treated as 1.6%.

Before the concentration of Si content becomes homogenization, as the diffusion time t is certain, and M and D are also certain constants, $\frac{M}{\sqrt{\pi D t}}$ is constant. So the decay factor of concentration $\exp\left(-\frac{x^2}{4Dt}\right)$ can be used to solve the diffusivity of Si in the samples. To facilitate the calculation, take logarithm on both sides of (2) and (3):

$$\ln\left(c(x,t_0)-A\right)=\ln\frac{M}{\sqrt{\pi D t_0}}-\frac{x^2}{4Dt_0} \qquad (4)$$

$$\ln\left(c(x,t_1)-A\right)=\ln\frac{M}{\sqrt{\pi D t_1}}-\frac{x^2}{4Dt_1} \qquad (5)$$

Based on the above equations, linear fitting concentration curves before and after annealing can be made. The results are as follows:

900℃ :

Before

$$\ln\left(c(x,t_0)-1.6\right)=2.30667-1.49E-3x^2$$

(6)

After

$$\ln\left(c(x,t_1)-1.6\right)=1.71038-4.686E-4x^2$$

(7)

950 C :

Before

$$\ln\left(c(x,t_0)-1.6\right)=2.29954-1.73E-3x^2$$

(8)

After

$$\ln\left(c(x,t_1)-1.6\right)=1.28815-2.718E-4x^2$$

(9)

1000 C :

Before

$$\ln\left(c(x,t_0)-1.6\right)=2.26455-1.77E-3x^2$$

(10)

After

$$\ln\left(c(x,t_1)-1.6\right)=1.27638-2.445E-4x^2$$

(11)

1050 C :

Before

$$\ln\left(c(x,t_0)-1.6\right)=1.81137-6.1565E-4x^2$$

(12)

After

$$\ln\left(c(x,t_1)-1.6\right)=1.20814-1.328E-4x^2$$

(13)

Table1 Effect of annealing temperature on diffusion of Si in samples

Samples	Annealing tem/℃	Diffusion timet_0/min	Diffusivity$D\times10^{13}$/m$^2\cdot$s^{-1}
(a)	900	27.526	1.016
(b)	950	11.184	2.15
(c)	1000	9.617	2.448
(d)	1050	16.500	4.102

Because the concentration distribution of the electro-deposition samples is supposed as the distribution when the thin film components diffuse for t_0, and the concentration distribution after annealing is the distribution when the thin film components diffuse for t_1, so we should make sure the occurrence of t_0. According to the slope of the fitting result before and after annealing, it is not difficult to see that the radio of t_0 and t_1 is inverse radio. Use k to stand for the slope of a line. Then $k_{before}/k_{after}= t_1/ t_0$ and t_1=(t_0+60)min. Put the value of t_0 into (4) and (5), and we can get the diffusivities of different annealing temperatures. The effects of annealing temperature on diffusion of Si in samples are shown in table 1, It is clear that the diffusion of

Si in the samples increases obviously, as is shown in article [6].

2.3 The analysis of sample structure and texture after annealing

The concentration distribution of sample in the process of electro-deposition has little influence on the diffusion of Si content. So, at a certain temperature, the annealing time of homogenizing annealing diffusion is mainly determined by the Si content in the electro-deposition silicon layer. Changing the parameter of the electro-deposition, the samples with different concentrations need to be determined the annealing time based on the Si content in the electro-deposition. According to the influence of the temperature on the diffusion of Si we get above, the study choose temperature at which higher diffusivity can be obtained to do the annealing experiment. The samples are analyzed after annealing. The result is shown in Fig 2.

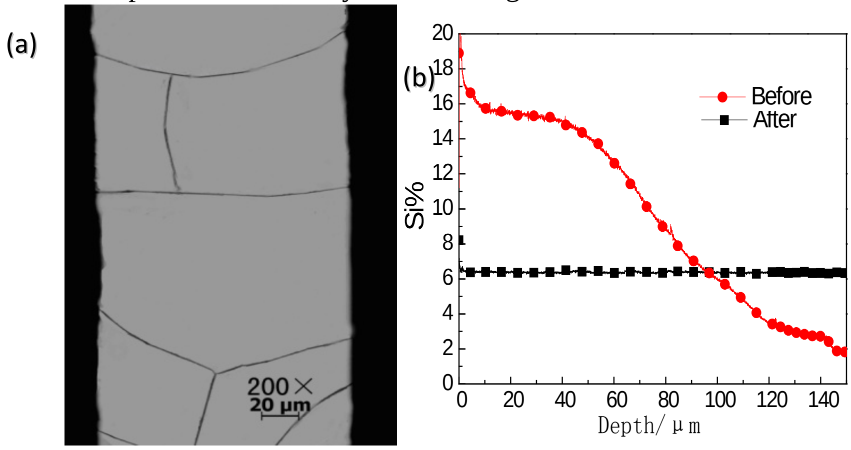

Figure 2 the metallographic photos of samples after the annealing and the Si concentration distribution metallographic photos (a) the metallographic photos (b)Si concentration distribution

As it is shown in Fig 2, the size of the crystal grains of the section in the samples are big, about 188.840μm, on the average of 1.5, which just falls into the best iron losses size range (100～200μm). The annealing process can improve the growth of crystal grains. The annealing temperature has great influence on the growing of the crystal grains. With the increase of the annealing temperature, the growing speed of the crystal grains increases. And after a certain period of time, coarse grains are generated. According to the relationship between the temperature and the diffusivity of Si content, the diffusivity of Si is bigger when the temperature is higher, the energy of the atom is bigger, and the diffusion is easier, too. High temperature annealing contributes to the diffusion of Si with high concentration from the surface into the inside of the samples. The concentration of the sample surface decreases. The depth of the diffusion increases. Finally the concentration of Si gets homogenization. At the temperature of 1000 C, 210min for annealing. the concentration tends to be evenly distributed, with the concentration of 6.3715%, close to 6.5%.

To get well-magnetic silicon steel, we need to improve the strength of the {100} and {110} texture, and decrease the strength of {111}[7-9]. It is mainly influenced by annealing conditions whether the texture after annealing the same as the texture after cold rolling. That is to say, it is mainly influenced by the annealing temperature and time. Fig 3 is the φ_2=45°sections of ODF for samples with different temperatures. It is known that after annealing, the textures in the sample mainly center on the {111}<112>, {111}<110> textures of γ fiber component, {100}<011>, {110}<490>, and {100}<001> textures of α fiber component, and {011}<100> Goss textures. Compared with the cold rolling sheet in 3(a), the density of the textures after annealing in γ fiber component decreases, in which the density of {111}<110> textures declines obviously. The components in cold rolling samples are mainly {111}<112>, {111}<110>, {100}<011>, {011}<100> and {112}<110>. The storages of every component, from strong to weak, are in turn {111}<112>, {111}<110>, {112}<110>, {100}<011>[10-11]. While annealing, nucleation and growth during Re-crystallization proceed in the {111} plane on the textures of higher storages {111}<112>、{111}<110>, while it is hard to form nucleation in the textures of low storages. So, the annealing textures priority in {111} plane. The grains in Goss texture {110}<001> gradually grows through consumption around {111} {111} orientation face in <112> texture components. When the annealing time reaches 210 min, the density of γ fiber component declines obviously. The density of Goss texture {110}<001> is high, about 6.20.

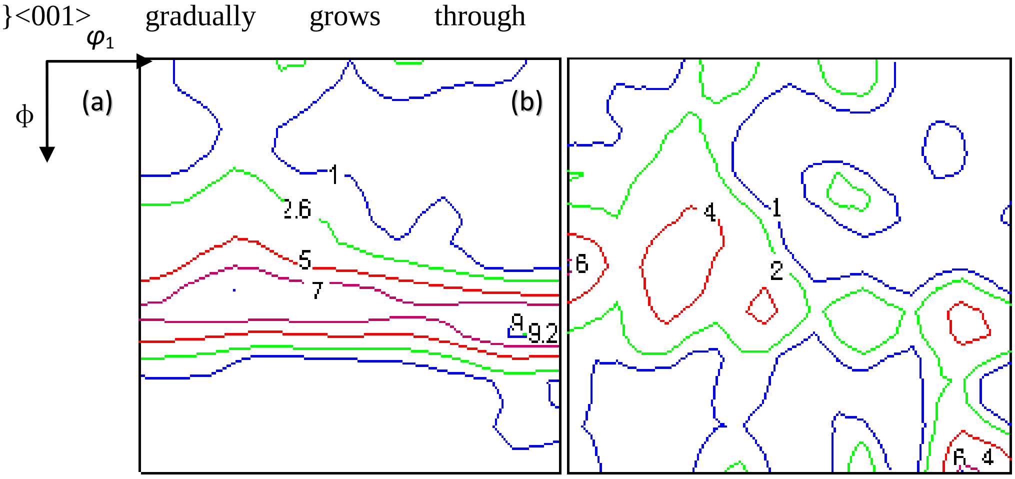

Figure 3 φ_2=45°sections of ODFs for sample(levels: 1, 2, 4, 5, 6, 9, 9.2)

3 Conclusions

The main annealing conditions are the influence of the change of temperature and time on the density of Si and the size of the grains, as well as the textures. The following conclusions can be made.

1) The Si content in surface layer decreases and tends to get homogenization. With the increase of the annealing temperature, the diffusivity of Si increases.

2) When the temperature gets to 1000 C, and annealing time is 210min, the size of the crystal grains is 188.840μm, falling into the best iron losses size range (100～200μm). The concentration of Si in the sample becomes homogenization, and gets thin silicon steel sheet with the Si content of 6.3715%, close to 6.5%.

3) The density in {100} and {110} is high, the density in {111} texture decreases.

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