# **The Effect of Heat-Treatment in a Magnetic Field or Under an Applied Stress on the Magnetic Properties of Silicon-Iron**

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The effects of heat-treatment in a magnetic field or under an applied stress have been studied in this investigation. Magnetic properties (magnetostriction and power loss in particular), measured along the rolling direction in grain-oriented silicon-iron were unaffected by magnetic annealing, but their stress-sensitivities were improved by annealing under tension.

Magnetic annealing was found to be effective in non-oriented silicon-iron and also in grain-oriented material if it was annealed with the field applied along directions other than the rolling direction.

The magnetic annealing results can be explained largely on the basis of the N6eI-Taniguchi theory of directional ordering of atom pairs. The changes obtained by annealing under stress showed that directional order only plays a minor part. The magnetic changes could be explained by assuming that during heat-treatment under stress a process of magnetostriction strain relief occurs, forming a residual internal stress.

Other alloys similar to silicon-iron showed no more response to magnetic annealing or annealing under stress than silicon-iron.

## **1. Introduction**

It has been known for many years that certain nickel-iron alloys are very susceptible to heattreatment in a magnetic field. Striking reductions in loss and magnetostriction have been reported by many workers and in a few cases the process is commercially important.

Few detailed studies of the effects of magnetic annealing or annealing under stress have been made on silicon-iron. This is mainly because initial results had shown only small improvements in properties compared with the nickeliron alloys.

Néel [1] and Taniguchi and Yamamoto [2, 3] independently developed similar theories which explain adequately most of the experimental results in nickel-iron. According to the Néel-Taniguchi theory, magnetic annealing, and the changes caused by annealing under stress, arise from a type of ordering of pairs of like atoms in a binary alloy. The ordered pairs of atoms are aligned either parallel or perpendicular to the magnetic field or stress direction.

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An investigation was carried out with these points in mind to see whether better conventional magnetic materials could be obtained by:

(i) Studying the effects of heat-treatment in a magnetic field or under an applied stress on the magnetic properties of oriented and nonoriented silicon-iron.

(ii) Examining the relevance of the Néel-Taniguchi theory to the experimental results obtained from silicon-iron.

(iii) Considering the extent to which other magnetic alloys (say silicon-iron with small additions of other elements) would respond to such treatment.

## **2. The Experimental Results**

2.1. Grain-oriented (Goss) and Non-oriented Silicon-Iron Annealed in a Magnetic Field

Grain-oriented  $3\frac{1}{4}$ % silicon-iron in the form of Epstein strips cut parallel to the rolling direction was annealed under various conditions in a longitudinal magnetic field. The power loss and magnetostriction were measured using an electronic wattmeter\* and a stereo ceramic transducer method [4] respectively.

The samples were annealed over a range of temperatures from 400 to  $900^\circ$  C and cooled at rates from 10 to  $100^{\circ}$  C h<sup>-1</sup>, with or without the presence of an external magnetic field. No change in properties could be detected after any of these heat-treatments.

Epstein strips of grain-oriented  $3\frac{1}{4}\%$  siliconiron were cut from a sheet at various angles to its rolling direction and given a stress relief anneal. They were next annealed in the presence of a 10 Oe field to study the effects of magnetic annealing along directions other than the rolling direction.

The optimum annealing condition was found to be to hold the samples at  $700^\circ$  C for 1 h and then cool at 300 $^{\circ}$  C h<sup>-1</sup> to 300 $^{\circ}$  C with the field still applied. Fig. 1 shows a typical curve of change in power loss produced after annealing the different strips. The greatest improvement occurs at about  $50^\circ$  to the rolling direction and even at  $90^\circ$  to the rolling direction the loss is reduced by 15% (this varied from 10 to 20% for different batches of samples). The corresponding effects of magnetic annealing on the dynamic magnetostriction (100 Hz component 11 kG) are shown in fig. 2.



*Figure 1* Effect of magnetic annealing on the total **power loss** (50 Hz, 11 kG) of Epstein strips of Goss-oriented  $3\frac{1}{4}\%$  silicon-iron cut at various angles to the rolling direction annealed in a longitudinal field.  $\bigcirc$ , before magnetic anneal;  $\times$ , after magnetic anneal.

Epstein samples of non-oriented  $2.7\%$  siliconiron were annealed in a magnetic field under the same conditions as the oriented material. Fig. 3 \* To be published.  $\dagger$  1 lb(f) in. -  $^2$  = 6894.76 N m<sup>-2</sup>. 464



*Figure 2* Variation of magnetostriction (100 Hz component, 11 kG) caused by annealing Epstein strips of Goss-oriented  $3\frac{1}{4}\%$  silicon-iron cut at different angles to the rolling direction in a longitudinal field.

shows the change in stress sensitivity of power loss and magnetostriction caused by the anneal. The reduction in loss is about  $7\frac{\nu}{6}$ , and the reduction in magnetostriction is about  $1 \times 10^{-6}$ at all stress levels (i.e.  $+800$  to  $-800$  lb(f) in.<sup>-2</sup>);<sup>†</sup> both are significant improvements.



*Figure 3* Change in stress sensitivity of power loss (13 kG, 50 Hz) and magnetostriction (100 Hz component, 13 kG) of 2.7% non-oriented silicon-iron caused by magnetic annealing in a longitudinal field of 11 A cm<sup>-1</sup>.

## 2.2, Grain-oriented and Non-oriented Silicon-iron Annealed Under Stress

Epstein strips of Goss-oriented material were cut from a sheet at various angles to its rolling direction and given a stress relief anneal. They were next annealed under longitudinal tension or compression for 1 h at 700 $\degree$  C and cooled at  $100^{\circ}$  C h<sup>-1</sup> to 300° C still under stress. Fig. 4 shows typical changes in power loss caused by annealing under tension and compression.

It can be seen that annealing with the tension applied along a direction greater than about  $55^\circ$ to the rolling direction reduces the loss, but below  $55^\circ$  the loss increases, and along the rolling direction itself it remains constant. The opposite occurs after annealing under compression.



*Figure 4* Changes in power loss (11 kG, 50 Hz) of Epstein strips cut at various angles to the rolling direction and annealed under stress, a, b, c, at 50 Hz; a', b', at 25 Hz; a,  $a'$ , annealing alone; b, b', 490 lb(f) in.<sup> $-2$ </sup> tension anneal; c, 380 lb (f) in. $-2$  compression annealed.

From fig. 4 it might appear that the magnetic properties along the rolling direction are unaffected by annealing under tension or compression. However, fig. 5 shows that there is an improvement in stress sensitivity along the rolling direction after heat-treatment under tension. The "shift" in the stress sensitivity curve can be explained in terms of a release of magnetostrictive strain during annealing causing a residual internal tension to be left after heattreatment. Similar shifts in stress sensitivity

curves of samples cut at other angles to the rolling direction were obtained. Samples heattreated under compression showed a shift of the curve as if a residual internal compression were formed.



*Figure 5* Change in stress-sensitivity of power loss and magnetostriction of an Epstein strip of  $3\frac{1}{4}\%$  Gossoriented silicon-iron cut along the rolling direction and annealed under a tension of 750  $1b(f)$  in.<sup> $-2$ </sup>.

Similar changes in stress sensitivity of loss and magnetostriction were obtained from nonoriented silicon-iron after annealing under tension. Fig. 6 shows a typical change due to annealing under tension. As with the oriented material heat-treatment under compression shifts the stress sensitivity curve as if an internal compression is produced.

The dependence of the change in power loss and magnetostriction on the magnitude of the tension applied during stress-annealing is shown in fig. 7. The optimum tension appears to be about 500 lb(f) in.<sup> $-2$ </sup> for both oriented and nonoriented material. The optimum annealing temperature was found to be about  $500^{\circ}$  C.

To investigate more fully the mechanism of annealing under stress, discs were etched from



*Figure 6* Change in stress sensitivity of power loss (13 kG, 50 Hz) and magnetostriction (13 kG, 100 Hz component) of 2.7% non-oriented silicon-iron caused by annealing under a tension of 750 lb(f) in.  $^{-2}$ .



*Figure 7* Variation in total **power loss and** magnetostriction caused by annealing 2.7% non-oriented siliconiron under different tensions.

Epstein strips of non-oriented silicon-iron which had been annealed in a magnetic field or under stress. Each sample was placed in a hightemperature torque magnetometer, a torque curve plotted, and the sample given a stress relief anneal and the torque curve repeated at room temperature. A stress relief anneal re-466

moved the effects of magnetic annealing or annealing under stress so any changes in torque curves would be due to the removal of any induced anisotropy. The sample which had been annealed in a magnetic field showed a torque change characteristic of a uniaxial anisotropy but the samples previously annealed under stress showed no change. This shows that as far as silicon-iron is concerned, atomic ordering is not the cause of the changes in power loss and magnetostriction produced by annealing under stress.

#### **3. Discussion of Results**

Magnetic annealing of grain-oriented  $3\frac{1}{4}\%$ silicon-iron has little practical importance except possibly where improvements are desired perpendicular to the rolling direction. In this case the loss can be reduced by between 10 and  $20\%$ and the magnetostriction by about 20 %.

Samples annealed under compression generally show a deterioration in magnetic properties. An Epstein sample of grain-oriented silicon-iron cut parallel to the rolling direction and annealed under a compression of 1000 lb(f) in. $-2$  showed a change in magnetostriction of from  $-0.35 \times$  $10^{-6}$  to  $+3.8 \times 10^{-6}$  and a  $10\%$  increase in loss. Thus it is important not to compress strips during annealing, accidentally.

Annealing under tension in the transverse direction in grain-oriented silicon-iron produces an improvement in loss, magnetostriction and stress sensitivity. Annealing under tension in the rolling direction improves the stress sensitivity only. This can however be a large improvement, for instance, a typical reduction in magnetostriction (caused by annealing under a tension of 780 lb(f) in.<sup>-2</sup>) was from  $+3 \times 10^{-6}$  to zero and the corresponding reduction in loss was  $9\%$ .

It has been known for some time that annealing under tension can be used to produce flatter sheet [5]. The catenary annealing process does this and improves the loss but the results given here show that it should also improve the stress sensitivity of the material whether a phosphate coating is applied during the anneal or not. (The higher the tension that can be applied without permanently straining the material the greater will be the shift in the stress-sensitivity curve and the better will be the improvement in loss and magnetostriction under compression.)

The fact that no change in torque is produced after annealing under stress shows that any anisotropy change is small compared with the change caused by magnetic annealing and the changes in loss and magnetostriction are almost entirely due to the apparent internal stress that is produced.

No change in stress sensitivity was found in silicon-free steel after magnetic annealing or heat-treatment under stress. According to the Néel-Taniguchi theory no change would be expected after magnetic annealing but the fact that no change was found after annealing under stress means that no magnetostrictive stress relief occurs either.

## **4. Further Experiments on the Relevance of the Néel-Taniguchi Theory to the Experimental Results**

The Néel-Taniguchi theory essentially states that magnetic annealing gives rise to a directional ordering causing a uniaxial anisotropy to be superimposed on the normal crystal anisotropy of a material. The most convenient method of detecting this type of anisotropy is by means of high-field torque curves. The changes expected in silicon-iron after magnetic annealing are only a few per cent of the total torque. For this reason a high-temperature torque magnetometer was necessary to heat-treat, magnetically anneal and measure the torque of samples accurately.

A typical measuring procedure was to plot a torque curve of a disc sample, heat it up to  $700^{\circ}$  C and hold for 1 h in a saturating magnetic field and cool at  $100^{\circ}$  C h<sup>-1</sup> to  $300^{\circ}$  C with the field still applied. The torque curve was replotted at room temperature and the change in torque represents the torque due to any induced anisotropy.

Fig. 8 shows the torque change caused by magnetic-annealing a sample of non-oriented



*Figure 8* Effect of magnetic anneal on the torque curves of non-oriented 2.7% silicon-iron, a, before magnetic **anneal;** b, after; c, change due to magnetic annealing. (The torque **is plotted against** the angle between the field direction and the magnetic annealing direction.)

2.7% silicon-iron. The difference in torque curves before and after heat-treatment is approximately of the form  $T = K_u \sin 2\theta$  where  $K<sub>u</sub>$  is a constant and  $\theta$  the angle between the magnetising direction and the magnetic annealing direction. In this case  $K_u$  is measured to be  $680$  J m<sup>-3</sup> and a calculated value based on the Taniguchi theory is  $280 \text{ J m}^{-3}$ . These agree quite well, considering the approximations necessary when calculating the theoretical value. According to the Taniguchi theory of directional ordering the induced anisotropy in the (1 10) or (001) plane caused by a field applied along a [1 11 ], [110] or [001] direction is of the form

$$
E = -K_{\rm u} \cos^2 \theta \tag{1}
$$

where  $\theta$  is the angle between the magnetising direction and the magnetic annealing direction. To calculate  $K<sub>u</sub>$ , the uniaxial anisotropy constant, a slightly modified version of Taniguchi's expression for  $K_u$  is used, i.e.

$$
K_{\rm u} = \frac{A \ N \ n^2 \ C_1 C_0 \ I_{\rm s}(T)^2 \ I_{\rm s}(T)^2}{kT' \ I_{\rm s}(0)^4} \qquad (2)
$$

where  $\vec{A}$  is a constant depending on the type of lattice structure and the direction of the annealing field,  $N$  is the number of atoms per unit volume,  $n$  is the concentration of solute atoms,  $k$  is Boltzmann's constant,  $T'$  is the annealing temperature,  $I_s(T)$  is the value of the saturation magnetisation at the measuring temperature  $T$ (similarly for the annealing temperature  $T'$  and  $0^{\circ}$  K).

The constants  $C_0$  and  $C_1$  are the values of  $C_{AA}$  +  $C_{BB}$  – 2 $C_{AB}$  at the measuring temperature and annealing temperature, respectively.  $C_{AA}$  etc. are the coefficients of pseudodipolar interaction between nearest atoms of the two types A and B (here iron and silicon). Approximate values for  $C_0$  and  $C_1$  can be found assuming. pure dipole-dipole coupling in which case  $C^2$  =  $10 kT<sub>c</sub>K<sub>1</sub>$ , where  $T<sub>c</sub>$  is the Curie temperature, and  $K_1$  is the first anisotropy constant [6].

The values of  $K<sub>u</sub>$  calculated from torque curves are compared with the theoretical values of  $K<sub>u</sub>$  for various alloys in the table below.

It will be noticed from the table that the 6.2  $\%$ silicon-iron sample would not appear to respond very much better to magnetic annealing than 3.2% material since the induced anisotropy is not much greater. However, an induced anisotropy of the size found in the  $6.2\%$  alloy will have a far greater effect on the magnetic properties than it would in the 3.2% material

Material	Anneal direction	Experimental $K_{\rm u}$ (J m <sup>-3</sup> )	Theoretical $K_{\rm u}$ (J m <sup>-3</sup> )
$2.6\%$ Goss-oriented single crystal of silicon-iron	10011	$\Omega$	0
	[110]	350	195
	[111]	560	390
$2.9\%$ Goss-oriented single crystal of aluminium iron	10011	0	$\Omega$
	[110]	460	270
	[111]	650	540
$6.2\%$ Goss-oriented silicon-iron single crystal	10011	0	$\Omega$
	[110]	290	460
	[111]	800	920
$2.7\%$ non-oriented silicon-iron	rolling direction	680	280

TABLE I Comparison between experimental and theoretical values of uniaxial anisotropy contents for some **silicon** and aluminium iron **alloys** 

because the crystal anisotropy and magnetostriction are much smaller.

The fact that ordering cannot play a significant part during stress-annealing of silicon-iron is borne out by consideration of Néel's equation for the induced anisotropy constant due to annealing under stress. The uniaxial anisotropy constant  $K_u$ , in this case is given by

$$
K_{\rm u} = \frac{3C_{\rm A}{}^2 C_{\rm B}{}^2 L_0 D_0 P}{x_0 \, RT' r_0 g} \tag{3}
$$

where P is the tension, g and  $x_0$  are shear and compressibility moduli,  $D_0/r_0$  is four times the deviation from Vegard's law and R is the gas constant. T is the annealing temperature and  $C_A$ and  $C_{\text{B}}$  are the concentrations of the A and B type atoms respectively.  $L_0$  is a constant depending on the coupling coefficients between A and B atoms.

It can be shown [7] that for silicon-iron  $K_u$  is theoretically negative if annealed under tension. This would produce a deterioration in power loss and magnetostriction in both oriented and non-oriented material. The improvement in properties obtained by annealing non-oriented silicon-iron under tension, and the fact that an improvement or deterioration may be obtained in the oriented material depending on the direction of the applied tension, show that the ordering process is completely overshadowed by some other process.

The changes in loss and magnetostriction in silicon-iron are in most cases similar whether annealed under tension or in a magnetic field. The experimentally induced anisotropy due to magnetic annealing was of the order of a few 468

hundred J  $m<sup>-3</sup>$  whereas it was undetectable after annealing under stress. Substituting the relevant values in equation 3 for annealing silicon-iron under stress gives a value of  $K<sub>u</sub>$  of the order of  $5$  J m<sup>-3</sup> which could not in itself affect the magnetic properties materially. Hence both the experimental results and theoretical estimates show that directional ordering is not the cause of the effect of annealing silicon-iron under stress.

## **5. Conclusions**

(i) Annealing in a magnetic field or under an applied stress can improve the power loss and magnetostriction of grain-oriented silicon-iron. Magnetic annealing, or annealing under tension, along the transverse direction can reduce the magnetostriction and power loss. This could be of some use for annealing the corners of transformer limbs, for instance. Magnetic annealing does not have any effect along the rolling direction, but the stress sensitivity of loss and magnetostriction can be improved by annealing under tension in the rolling direction. This occurs in both oriented and non-oriented material. It shows that a final catenary type anneal or thermal flattening could be of benefit in the steel-producing process provided no anneal is carried out afterwards.

(ii) The results obtained are quite repeatable; the Néel-Taniguchi theory gives an explanation of the magnetic annealing results in terms of directional ordering. Annealing under stress, however, causes effects as if an internal stress is produced by magnetostriction strain release.

(iii) Other materials similar in composition to

silicon-iron were not found to be any more susceptible to annealing, under stress or in a magnetic field. These included two tertiary alloys and silicon-free steel.

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