Frequency behaviour of SPD treated soft magnetic materials

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Abstract. The effect of SPD treatment on the hysteresis properties of selected soft magnetic materials such as Fe3%Si and Fe17%Co steel is presented. The enhancement of the coercivity due to the mechanical deformation can be explained by the magnetoelastic energy. The frequency dependence of the coercivity and of the losses can be described using an eddy current based model.

1. Introduction

Soft magnetic materials are characterised by the shape and the most important points (coercivity, remanence, losses) of their hysteresis loop. Figure 1 shows for a comparison the typical hysteresis loops of soft and hard magnetic materials.

![Figure 1. Typical hysteresis loop of soft and hard magnetic materials.](image)

Generally soft magnetic materials are formed by 3d metals such as Fe, Co or Ni respectively 3d based alloys. Here well-known examples are Fe-Si, Fe-Si-Al, Fe-Ni etc (see e.g. [1]).

In general, technically useful soft magnetic materials should exhibit the following intrinsic properties at room temperature: a high saturation magnetization, a low anisotropy and a low magnetostriction. Additionally extrinsic properties such as low coercivity, high permeability and low losses are important. The extrinsic properties are determined by the microstructure such as grain size, different phases, grain boundaries and internal stresses, which can be influenced by the heat treatment but also by plastic deformation.
In this paper, the magnetic properties of some important soft magnetic metallic materials such as Fe-3%Si as well as a standard steel will be given and discussed. In many technical applications these materials are produced or used with stresses added. In order to investigate the effect of stresses on the hysteresis properties, the samples were produced by applying a so-called SPD (Severe Plastic Deformation) treatment [2]. The hysteresis properties of SPD-treated soft magnetic materials were investigated in detail within the PhD thesis of N. Mehboob [3].

2. Experimental

**Samples:** In the last years, the magnetic properties (frequency dependence of the hysteresis loop, losses) on all kind of soft magnetic materials, such as pure Fe, pure Ni, steels, Fe3%Si and Fe6.5%Si [1,3] but also amorphous and nanocrystalline materials [4,5] were investigated. Within this review we will focus only on Fe-based alloys, i.e. a Fe3%Si (electro-steel) and a Fe-Co steel.

All bulk samples were studied in an “undeformed” state but also after a so-called SPD (Severe Plastic Deformation) treatment which is nowadays a well established method to produce nanocrystalline bulk materials [2]. Specimens have been disc-shaped to a diameter of 10 mm, and from each material a set of four samples was produced. These samples were deformed by High Pressure Torsion (“HPT” being one of the most effective SPD methods [6], see also below) at liquid nitrogen temperature (77 K), ambient temperature (RT = 298 K), and 723 K corresponding to 0.4 $T_{\text{hom}}$ ($= T_{\text{HPT}} / T_{\text{melting}}$) under a force of 500 kN which corresponds to a pressure of about 6 GPa. With the rotation tools, three complete turns were done, thus achieving an average torsional shear strain of about $\gamma = 80$. One sample from each set was kept undeformed as a reference. Ring-shaped samples were made from the periphery of the thin HPT-deformed discs, which are in the saturation state of deformation and microstructure, and from magnetic point of view the best choice for hysteresis measurements. The HPT processing parameters are given in Table 1.

Figure 2 shows the principle of HPT setup where a high pressure combined with torsion at different temperatures can be applied. The drawing is from the facility used in this work, which is available at the Erich Schmid Institute, Austrian Academy of Sciences, Leoben; Austria with two systems applying at maximum force of 500 kN, and 4MN, respectively. This treatment can be performed between 77K and 800 K. In the periphery of sample (see e.g. Figure 7), a maximum of grain refinement is achieved. Summarizing, this method can cause nanocrystallization [7] as well as a strongly deformed state [8].

![Figure 2. Schematic view of the used HPT equipment for hot (left) and cold (right) processing.](image)
Table 1: HPT parameters as used for pure Fe-3 wt%Si and Fe-17wt %Co samples, $T_{\text{HPT}}$ - HPT treatment temperature; n-number of turns; t - thickness of sample before HPT; $d_o$-outer diameter; $d_i$-inner diameter of ring-shaped samples. The pressure was about 6 GPa (500kN over a sample area of 80 mm²), the average torsional shear strain was about 80.

| Sample    | $T_{\text{HPT}}$ | N  | t [mm] | $d_o$ [mm] | $d_i$ [mm] |
|-----------|------------------|----|--------|------------|------------|
| Fe3%Si    | 0                | 0  | 0,81   | 7,053      | 5          |
| Fe3%Si    | RT               | 3  | 0,638  | 7,01       | 5          |
| Fe3%Si    | 723 K            | 3  | 0,637  | 7,026      | 5          |
| Fe3%Si    | 77 K             | 3  | 0,653  | 6,91       | 5          |
| Fe17%Co   | 0                | 0  | 0,781  | 7,053      | 5          |
| Fe17%Co   | RT               | 3  | 0,64   | 7,01       | 5          |
| Fe17%Co   | 723 K            | 3  | 0,657  | 7,026      | 5          |
| Fe17%Co   | 77 K             | 3  | 0,653  | 6,91       | 5          |

3. Hysteresis measurement

For measuring the frequency dependence of the hysteresis loop (at room temperature) we used a fully automated hysteresigraph which was described in [9, 10] - see figure 3. Its main part is a standard PC with a National Instrument measurement card (NI 6120). The measuring electronics for generating the magnetic field consists of an external power amplifier (KEPCO) for generating the magnetizing current. The whole measurement is handled by a LabView program. The advantages of such a system are: i) the current $I(t,f)$ can be chosen freely as e.g. triangular, sinusoidal one etc, ii) averaging over many loops which improves the signal - noise ratio, iii) automatic demagnetization of the sample, iv) frequency dependent measurements, v) temperature dependent measurements. The software was recently improved in order to measure under constant dB/dt conditions which is necessary to determine the true losses [11]. The hysteresis measurements are usually performed at room temperature; however, other temperatures (as e.g. -30°C or + 100°C) are also possible. All samples were demagnetized before each hysteresis measurement.

Figure 3. Block diagram of the Labview-automated hysteresis measurement system for ring-shaped samples
The frequency dependence of the hysteresis loop was measured on ring-shaped samples at room temperature applying a triangular field \( H(t) \). Using a triangular field allows to determine the relative permeability \( \mu_r(H) \) directly from measurements of minor loops using formula (1):

\[
\mu_r(H) = \frac{1}{\mu_0} \frac{\Delta B(H)}{\Delta t f H_{\text{max}}}
\]

4. Results and Discussion

First we want to summarise the intrinsic magnetic properties of pure Fe and compare with Fe3%Si – see table 2. However, the hysteresis properties such as the coercivity, the remanence etc. depend - beside the intrinsic properties - mainly on the actual microstructure such as grain size, defects etc.

|        | \( M_s \) (T) | \( K_1 \) (J/m\(^3\)) | \( K_2 \) (J/m\(^3\)) | \( \Delta M_\text{ps} \) (ppm) | \( \Delta M_\text{ps} \) (ppm) | Density (g/cm\(^3\)) |
|--------|---------------|------------------------|------------------------|----------------------------|----------------------------|------------------------|
| Fe     | 2.15          | 4.9.10\(^4\)          | 1.1.10\(^4\)          | 21                        | -21                       | 9.61                   |
| Fe3%Si | 2.0           | 3.6.10\(^4\)          | -                      | 23                        | -4                        | 48.0                   |

Table 2. Room temperature data of the saturation magnetization \( M_s \), the crystal anisotropy \( K_1 \) and the magnetostriction \( \Delta M_\text{ps} \) of pure Fe compared with the data of Fe3%Si [13].

The hysteresis loops of undeformed and HPT-deformed Fe, Fe-3 wt% Si, Fe-6.5 wt% Si and Fe-17 wt%Co (P800) samples were measured. Figure 4 shows - as an example - measurements of Fe3%Si which were performed at room temperature in the frequency range of 0.25 Hz to 1 kHz. The maximum average applied field was about 20000 A/m. It can be seen from the width of the hysteresis loops that the coercivities \( H_c \) increased significantly with frequency. This behaviour can be simply described using an eddy current based formula (2) of the type:

\[
H_c(f) = a + b \sqrt{f} + c f
\]

The strong frequency dependence of the coercivity \( H_c \) at high frequencies is understood in iron (which is a well conductive material), due to the increasing effect of eddy currents induced in the material with increasing frequency [12].

It is well known that the addition of non-magnetic silicon lowers the saturation flux but significantly decreases the coercivity. For example the coercivity is reduced up to factor 2.5 with 3 wt%Si as compared to pure undeformed iron whereas the electrical resistivity of pure iron increases about 4.5 times with the addition of 3 wt%Si and 8 times with 6.5 wt%Si [13], thereby reducing the eddy currents. Moreover with increasing Si content also the magnetocrystalline anisotropy constant \( K_1 \) [12] increases which is obvious from the width of the hysteresis loops.

The coercivity of all HPT-deformed samples at low frequencies is considerably high as compared to undeformed material: although the grain size is reduced to nano-scale due to the SPD deformation, it is still larger than the exchange coupling length. Moreover, the SPD deformation increases the level of microstresses \( \sqrt{\tau} \) as well as of the number of dislocations and other lattice defects which leads to domain wall pinning. Thus the deformation increases the magnetoelastic energy \( \Delta E \) which results in an increase of the (static) coercivity – see formula (3).

\[
H_c = \frac{\sigma \Delta + \langle \kappa \rangle}{\mu_0 M_s}
\]
However, at higher frequencies, the effect of eddy current is comparatively smaller in HPT-deformed samples as compared to undeformed ones because the deformation induced dislocations/lattice defects also enhance the electrical resistivity and reduce the permeability of the material.

Figure 4 shows as examples the frequency dependent hysteresis loops as measured on undeformed bulk polycrystalline Fe3%Si and on a RT-SPD processed sample.

![Figure 4](image)

**Figure 4.** Frequency dependence of the at room temperature measured hysteresis loop of undeformed (left picture) and of RT-SPD deformed (right picture) Fe3%Si.

$H_c(f)$ of FeSi shows a nonlinear relation between the coercivity and the frequency, indicating the importance of eddy currents within the dynamic process (see figure 5). $H_c(f)$-curves for deformed samples are less steep as compared to undeformed samples. The slope of the $H_c(f)$-curve is reciprocally proportional to the square root of the electrical resistivity of the material. This indicates that the resistivity of the material increases with deformation which is in line with the fact that deformation induced dislocations and/or lattice defects enhance the resistivity. Figure 6 shows the frequency dependence of the losses of all SPD treated Fe3%Si samples. The applied field $H(t)$ was triangular. Table 3 shows the initial and the maximum permeability (calculated from formula (1)), the coercivity extrapolated to zero frequency, as well as the ratio $B_r/B_{max}$. This ratio should be about 0.8 for an isotropic cubic material. The fact that all values are significantly below 0.8 can be explained assuming a strong induced texture. Applying formula (2) gives the fit parameters $(a,b,c)$ shown in table 3 and also achieves the results of a loss analysis using formula (4). This formula is based on a loss separation usual for Fe-Si transformer sheets [14]

\[
(BH) = W_{tot} = W_h + W_{class} + W_{exc} = W_h + B_2 f + B_1 f^{0.5} 
\]

\[
W_{tot} = W_h + CB_{max}^{1.5} f^{0.5} + a \frac{A \pi^2 B_{max}^2}{6 \rho} f = W_h + C B_{max}^{1.5} f^{0.5} + a W_c f 
\]

where $W_h$ represents the frequency independent hysteresis losses, and $B_1$, $B_2$ are fitting parameters. $A(m^2)$ is the cross section of the sample, $B_{max}$ gives the maximum induction of the material and $\rho$ is the specific electrical resistivity; $a$ describes a constant which is determined by the shape of the driving field. The excess losses can be describe according to Bertotti [14] with a formula which contains mainly a hypothetic field $V_0$ which is an important parameter for the damping process:

\[
W_{exc} = C. B_{max}^{1.5} \sqrt{f} = 8 \sqrt{G A V_0 \rho B_{max}^{1.5}} \sqrt{f} 
\]

The value of $V_0$ can be estimated by using the parameter $B_1$, the specific electrical resistivity of the material and $B_{max}$. 

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5. Fe-Co steel

In this material, all deformation states processed at different HPT temperatures correspond to a torsional shear strain of \( \approx 75 \). Crystallite sizes were extremely decreased in the highly deformed states. Deformed microstructures are shown in figure 7 following HPT at 723, 293 and 77 K, respectively [15]. After HPT the mean crystallite size as calculated by the area fraction method decreased from 270 to 83 and 72 nm after HPT deformation at 723, 293 and 77 K, respectively. By comparing the microstructures of Fe-17 wt% Co in figure 7 (b,c), one can clearly see that both the grain size and the grain shape have strongly changed.

Table 3: Initial permeability \( \mu_0 \), maximum permeability \( \mu_{\text{max}} \) determined at the field \( H(.,.) \), zero frequency coercivity \( H_c \) (A/m), and the ratio \( B_r/B_{\text{max}} \). Polynomial fit parameters \( (a,b,c) \) of the frequency dependence of the coercivity for all HPT treated Fe3%Si samples at RT. Fit parameters of the frequency dependence of the losses \( (W_h, B_1, B_2) \) using formula (4).

| Fe3%Si | \( \mu_0 \)  | \( \mu_{\text{max}} \) at \( H(A/m) \) | \( H_c \) (A/m) | \( a \) | \( b \) | \( c \) | \( W_h \)  | \( B_1 \)  | \( B_2 \)  | \( aB_2 \) | \( V_0 \) |
|--------|-------------|-----------------|----------------|---|---|---|---|---|---|---|---|
| Undef  | 1000        | 1000(115)       | 0.15           | 45 | 35 | 1,3 | 86 | 146 | 11 | a.35 | 244 |
| RT     | 290         | 1000(710)       | 0.23           | 659| 48 | 1,8 | 4392| 312 | 13 | a.26 | 1537|
| 723 K  | 120         | 900(920)        | 0.7            | 830| 41 | 1,9 | 4393| 126 | 13 | a.25 | 258 |
| 77 K   | 220         | 650(422)        | 0.4            | 552| 25 | 2   | 973 | 176 | 12 | a.25 | 503 |

Figure 5. \( H_c(f) \) of SPD treated Fe3%Si.

Figure 6. Frequency dependence of the losses of SPD treated Fe3%Si.
Figure 7. Grain size maps for Fe-17 wt% Co alloy corresponding to HPT temperatures (a) 723 K, (b) 293 K and (c) 77 K [15].

Figure 8 shows for comparison the frequency dependence of the hysteresis loops as measured on undeformed Fe-17wt%Co and on at RT deformed material. Note that the coercivity here is significantly higher than that of Fe3%Si. Figure 9 (left picture) shows the frequency dependence of the coercivity as obtained for all SPD treated steel samples. Figure 9 (right picture) shows the frequency dependence of the losses as obtained for all SPD treated steel samples.

Figure 8. Frequency dependent hysteresis loops of Fe-17wt%Co (a) undeformed, (b) HPT treated at RT
Figure 9. The frequency dependence of the coercivity (left picture) and the losses (right picture) of all HPT-treated Fe-17 wt% Co samples.

Table 4. Results of analysing the frequency dependence of the hysteresis loop (coercivity) applying formula (2) as well as the losses using formula (4), as follows:

| Samples | a   | b   | c | Wb | B1  | B2  | aB2 | V0 |
|---------|-----|-----|---|----|-----|-----|-----|----|
| Undef   | 372 | 63  | 1 | 1283 | 132 | 10  | a.296 | 23 |
| RT      | 1533| 22  | 2 | 7728 | 44  | 14  | a.215 | 4  |
| 723 (K) | 2510| 57  | 1 | 6 | 13760| 315 | 11  | a.121 | 168|
| 77 (K)  | 1379| 31  | 1 | 8 | 4150 | 108 | 10  | a.245 | 19 |

6. Conclusion
Severe plastic deformation (SPD treatment) generally causes an increase in the static coercivity as well as in the hysteresis losses $W_b$. In particular, the plastic deformation increases the magnetoelastic energy which is responsible for all effects on the hysteresis loop. The frequency dependence of the coercivity as well as of the losses can be described with the same type of formula which is based on the idea that there are two dynamic contributions, one which comes from the classical eddy currents and a second which arises from the movement of the domain walls (excess losses).

7. References
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