



# Magnetic properties of $\text{Fe}_{76}\text{X}_2\text{Si}_8\text{B}_{14}$ (X=Al, Cr, Mo) amorphous alloys

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## ABSTRACT

**Purpose:** The idea of the paper is to study the influence of thermal annealing and alloying additions on magnetic properties, optimization and crystallization processes in  $\text{Fe}_{76}\text{X}_2\text{Si}_8\text{B}_{14}$  (X=Al, Cr, Mo) amorphous alloys.

**Design/methodology/approach:** For annealed samples (1 h,  $T_a$  ranging from 300 K to 800 K) at room temperature magnetic permeability was measured by applying Maxwell-Wien bridge (frequency about 1030 Hz and magnetic field  $H=0.5$  A/m). Magnetostriction coefficients – parallel and perpendicular were determined by applying infra-red magneto-dilatometer. Magnetization in saturation versus temperature was measured by making use of magnetic balance (field 0.5 T).

**Findings:** It was shown that alloying additions in the examined alloys cause a decrease of the Curie temperature, an increase of magnetic permeability and magnetization in saturation. The observed ESMP (enhancement of soft magnetic properties) effect in the examined alloys can be attributed to the so-called relaxed amorphous phase free iron nanograins. It was shown that parallel and perpendicular magnetostriction coefficients depend on annealing temperatures which means that these quantities are sensitive on free volume content.

**Research limitations/implications:** The obtained results are a part of a broad area of examinations devoted to establishing of the influence of different alloying additions and thermal annealing on soft magnetic properties of amorphous alloys obtained by melt spinning technique.

**Practical implications:** The examined alloys belong to a modern group of soft magnetic materials, which can be used as core transformers, magnetic sensors, shields of magnetic etc. The obtained results may be used for preparing soft magnetic ribbons for specific applications.

**Originality/value:** The originality of the paper lies in examination of the influence of free volume content on magnetostriction coefficients.

**Keywords:** Amorphous materials; Soft magnetic properties; Magnetostriction coefficient; Crystallization

## MATERIALS

### 1. Introduction

Amorphous alloys based on iron obtained by melt-spinning technique are very interesting as soft magnetic materials. Many

properties of these alloys are superior to those of the conventional alloys with the same chemical composition. Moreover, some properties of these materials (like magnetic permeability, coercive field) can be significantly enhanced by applying a suitable

thermal annealing at temperatures close to the crystallization temperature. This enhancement of soft magnetic properties (ESMP) effect is usually explained by: i) formation of a nanocrystalline phase, i.e. by formation of iron nanograins:  $\alpha\text{Fe}$  or  $\alpha\text{Fe}(\text{Si})$  in amorphous matrix [1-8] or ii) annealing out of free volume and internal stresses leading to formation of the so-called relaxed amorphous phase free of iron nanograins [9-11]. In [9-11] it was concluded that the observed enhancement effect is due to formation of small iron clusters with different magnetic order.

In general, magnetic properties strongly depend on chemical composition of this kind of alloys. Even small amount of alloying additions can drastically change magnetic properties, the course of structural relaxation and crystallization processes. Therefore the aim of the present paper is to study the influence of thermal annealing and alloying additions on magnetic properties (magnetization in saturation, magnetic permeability, the Curie temperature), optimization (the optimization annealing temperature), and crystallization processes (the crystallization temperature) in  $\text{Fe}_{76}\text{X}_2\text{Si}_8\text{B}_{14}$  ( $\text{X}=\text{Al}, \text{Cr}, \text{Mo}$ ) amorphous alloys.

## 2. Experiment procedure

Experiments were carried out for amorphous alloys  $\text{Fe}_{78}\text{Si}_8\text{B}_{14}$ ,  $\text{Fe}_{76}\text{Al}_2\text{Si}_8\text{B}_{14}$ ,  $\text{Fe}_{76}\text{Cr}_2\text{Si}_8\text{B}_{14}$  and  $\text{Fe}_{76}\text{Mo}_2\text{Si}_8\text{B}_{14}$  obtained by melt spinning technique in the form of strips of thickness and width of about 20 - 30  $\mu\text{m}$  and 10 mm, respectively. As quenched ribbons were annealed for 1 hour at temperatures  $T_a$  ranging from 300 K to 800 K. For annealed samples at room temperature magnetic permeability  $\mu$  was measured by applying Maxwell-Wien bridge working at frequency about 1030 Hz and magnetic field  $H=0.5$  A/m.

Magnetostriction coefficients – parallel ( $\lambda_{||}$ ) and perpendicular ( $\lambda_{\perp}$ ) were determined also at room temperature (infra-red magneto-dilatometer working with a resolution of about 10 nm) for samples 5 cm in length using magnetic field oriented parallel or perpendicular to the measuring strain direction, respectively. From these measurements  $\lambda_s$  – saturation magnetostriction (anisotropic) and  $\omega$  – the so-called volume magnetostriction (isotropic) were calculated according to [12-13]:

$$\lambda_s = \frac{2}{3}(\lambda_{||} - \lambda_{\perp}) \quad \text{and} \quad \omega = \lambda_{||} + 2 \cdot \lambda_{\perp} \quad (1)$$

In order to determine the Curie temperature  $T_C$  and the primary crystallization temperature  $T_x$  for the as quenched ribbons magnetization in saturation  $\mu_0 M_s$  versus temperature (300 – 1000 K, linear heating rate 5 K/min) was measured by making use of magnetic balance (field 0.5 T).

## 3. Results

Figs. 1 and 2 show parallel and perpendicular magnetostriction coefficients versus applied magnetic field  $\mu_0 H$ . The influence of alloying additions on both magnetostriction coefficients is well demonstrated.

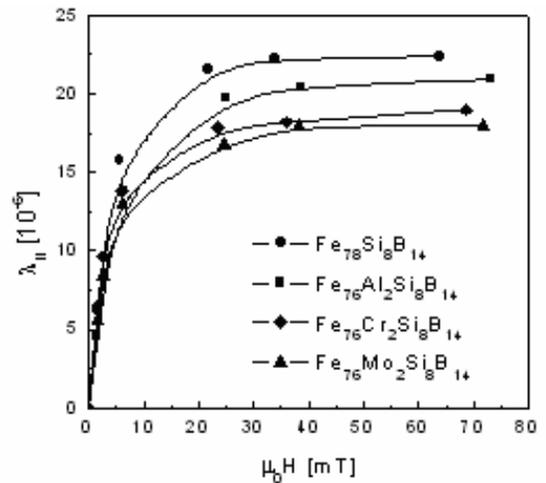


Fig. 1. Parallel magnetostriction coefficient  $\lambda_{||}$  versus magnetic field  $\mu_0 H$  for all examined alloys

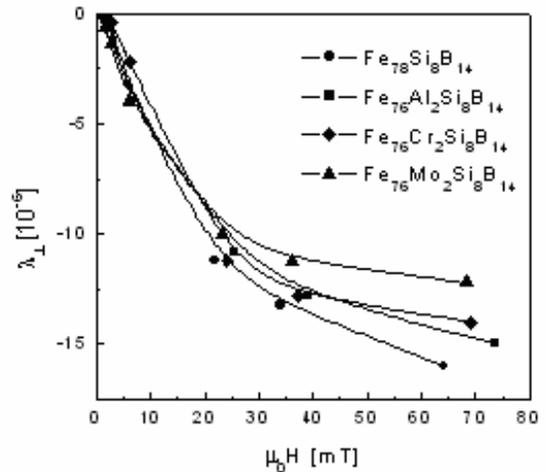


Fig. 2. Perpendicular magnetostriction coefficient  $\lambda_{\perp}$  versus magnetic field  $\mu_0 H$  for all examined alloys

Figs. 3-6 show parallel magnetostriction coefficient  $\lambda_{||}$  and magnetic permeability  $\mu$  determined for all examined alloys versus the 1-h annealing temperature  $T_a$ . In all cases permeability remains approximately constant up to  $T_a=520$  K, then increases with annealing temperature up to the maximum positioned at the so-called 1-h optimization temperature  $T_{op}$ . For  $T_a > T_{op}$  permeability drastically decreases. Parallel magnetostriction coefficient remains approximately constant up to  $T_a=520$  K, decreases with annealing temperature up to  $T_{op}$  and increases for  $T_a > T_{op}$ .

Fig. 7 shows magnetization in saturation  $\mu_0 M_s$  versus temperature  $T$  obtained for  $\text{Fe}_{78}\text{Si}_8\text{B}_{14}$  and  $\text{Fe}_{76}\text{Mo}_2\text{Si}_8\text{B}_{14}$  alloys. It can be seen that in both cases magnetization decreases with temperature up to the Curie point of amorphous phase. At higher temperatures the examined material is in paramagnetic state and magnetization is almost zero. For  $T > 700$  K (or 750 K for the  $\text{Fe}_{78}\text{Si}_8\text{B}_{14}$ ) magnetization in saturation increases due to formation of iron crystallites. Similar results were obtained for the other examined alloys.

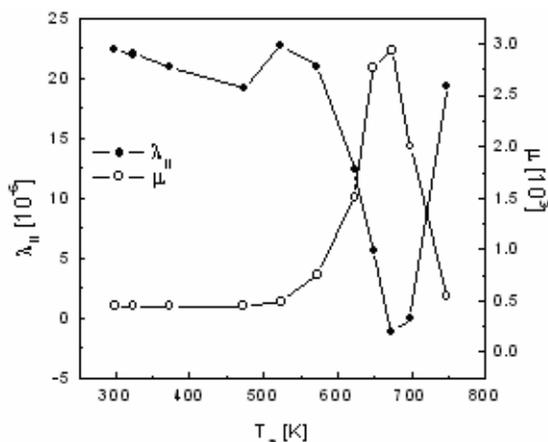


Fig. 3. Magnetic permeability  $\mu$  and parallel magnetostriction coefficient  $\lambda_{||}$  versus annealing temperature  $T_a$  for  $\text{Fe}_{78}\text{Si}_8\text{B}_{14}$  alloy

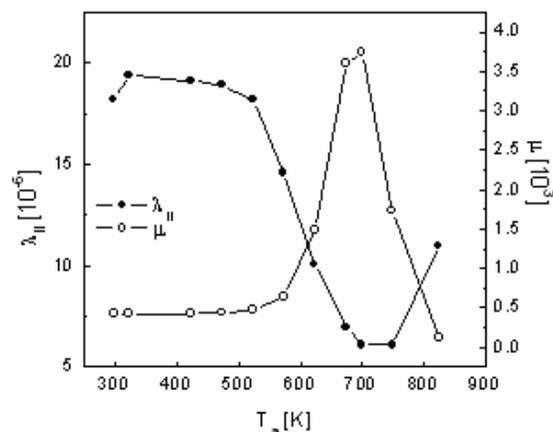


Fig. 6. Magnetic permeability  $\mu$  and parallel magnetostriction coefficient  $\lambda_{||}$  versus annealing temperature  $T_a$  for  $\text{Fe}_{76}\text{Mo}_2\text{Si}_8\text{B}_{14}$  alloy

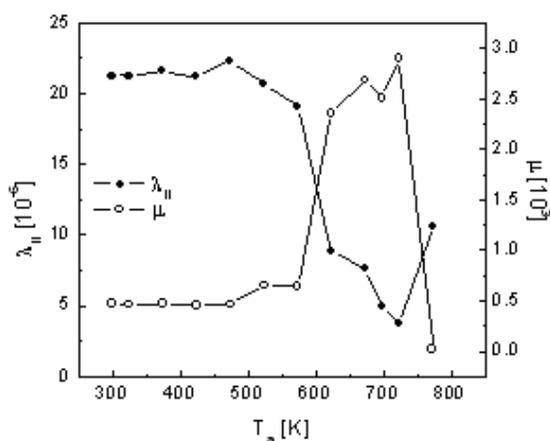


Fig. 4. Magnetic permeability  $\mu$  and parallel magnetostriction coefficient  $\lambda_{||}$  versus annealing temperature  $T_a$  for  $\text{Fe}_{76}\text{Al}_2\text{Si}_8\text{B}_{14}$  alloy

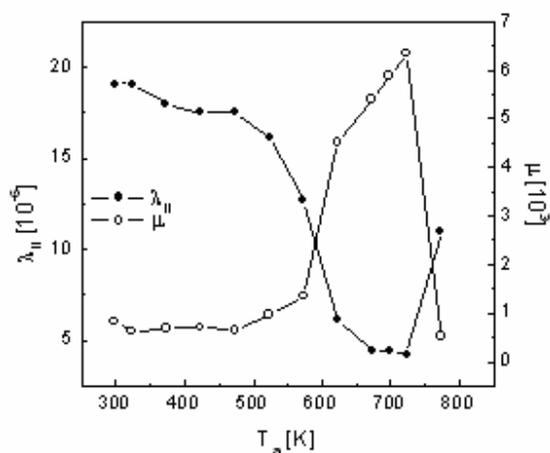


Fig. 5. Magnetic permeability  $\mu$  and parallel magnetostriction coefficient  $\lambda_{||}$  versus annealing temperature  $T_a$  for  $\text{Fe}_{76}\text{Cr}_2\text{Si}_8\text{B}_{14}$  alloy

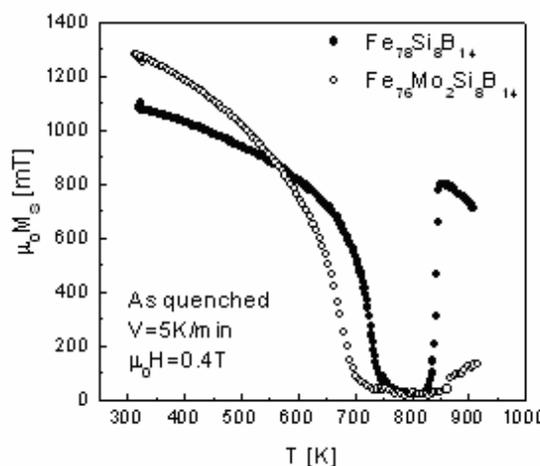


Fig. 7. Magnetization in saturation  $\mu_0M_S$  versus temperature  $T$  for  $\text{Fe}_{78}\text{Si}_8\text{B}_{14}$  and  $\text{Fe}_{76}\text{Mo}_2\text{Si}_8\text{B}_{14}$  alloys (heating rate 5 K/min)

#### 4. Discussion and conclusions

Magnetic parameters of the examined alloys determined in the present paper – the Curie temperature  $T_C$ , magnetization in saturation  $M_S$ , magnetic permeability  $\mu$  and magnetostriction coefficients  $\lambda_{||}$ ,  $\lambda_{\perp}$  are listed in Table 1 where the primary crystallization temperature  $T_X$  and the 1-h optimization annealing temperature  $T_{op}$  are also included.

From Figs. 3-6 and Table 1 it can be seen that all alloying additions cause a shift of the optimization maximum  $\mu(T_a)$  into higher temperatures in relation to the  $\text{Fe}_{78}\text{Si}_8\text{B}_{14}$  base alloy – molybdenum by about 25 K, chromium and aluminum by about 50 K. This means that the optimization effect strongly depends on chemical composition – the highest value was obtained for the  $\text{Fe}_{76}\text{Cr}_2\text{Si}_8\text{B}_{14}$  alloy (permeability increases more than 7 times in relation to the as quenched sample). Let notice that an increase of magnetic permeability corresponds to a decrease of parallel magnetostriction coefficient. Fig. 8 shows different

magnetostriction coefficients - i.e. parallel, perpendicular, saturation and volume (calculated according to equation (1)) plotted versus annealing temperature (up to 1-h optimization annealing temperature) for the  $Fe_{76}Cr_2Si_8B_{14}$  alloy. All coefficients  $\lambda_{||}$ ,  $\lambda_{\perp}$ ,  $\lambda_S$  and  $\omega$  remain approximately constant up to  $T_a=525$  K (just as magnetic permeability, see Figs. 3-6). For higher annealing temperatures  $\lambda_{||}$ ,  $\lambda_{\perp}$  and  $\omega$  decrease with increasing  $T_a$ . It means that these quantities are sensitive on free volume content. In contrast to this  $\lambda_S$  remains independent on annealing temperature. Similar effect was observed for the other examined alloys.

Table 1.

The Curie temperature  $T_C$ , the primary crystallization temperature  $T_X$ , the optimization annealing temperature  $T_{op}$ , magnetization in saturation  $\mu_0 M_S$ , magnetic permeability  $\mu$ , magnetostriction coefficients  $\lambda_{||}$ ,  $\lambda_{\perp}$  for the examined alloys

Alloy	$T_C$ [K]	$T_X$ [K]	$T_{op}$ [K]	$\mu_0 M_S$ [mT]	$\mu_{op}$	$\lambda_{  }$ [ $10^{-6}$ ]	$\lambda_{\perp}$ [ $10^{-6}$ ]
$Fe_{78}Si_8B_{14}$	730	840	675	1100	2950	22.5	-16
$Fe_{76}Al_2Si_8B_{14}$	725	845	725	1280	2900	21	-15
$Fe_{76}Cr_2Si_8B_{14}$	660	840	725	1340	3750	19	-14
$Fe_{76}Mo_2Si_8B_{14}$	675	865	700	1280	6350	18	-12

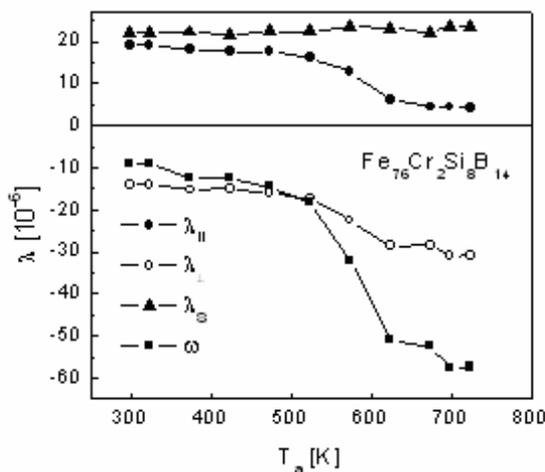


Fig. 8. Magnetostriction coefficients - parallel  $\lambda_{||}$ , perpendicular  $\lambda_{\perp}$ , saturation  $\lambda_S$  and volume  $\omega$  plotted versus the annealing temperature  $T_a$  for the  $Fe_{76}Cr_2Si_8B_{14}$  alloy

From Table 1 one can conclude also that all alloying additions cause an increase of magnetization in saturation at room temperature. The highest value of  $\mu_0 M_S$  was obtained for the  $Fe_{76}Cr_2Si_8B_{14}$  alloy. Molybdenum and chromium as alloying additions cause a decrease of the Curie temperature (position of the inflection point of  $M_S(T)$  curve) while aluminum does not influence  $T_C$  in relation to the  $Fe_{78}Si_8B_{14}$  base alloy. It is also clear that both aluminum and chromium do not influence the crystallization temperature  $T_X$  (position of the maximum of  $d(\mu_0 M_S)/dT$  curve). In contrast to this molybdenum causes a shift

of nanocrystallization into higher temperatures which means that Mo atoms cause a slowing down of diffusion processes leading to crystallization (nanocrystallization). It seems that the different behavior of Cr (or Al) and Mo atoms in the  $Fe_{78}Si_8B_{14}$  base alloy can be explained by differences in atomic radii and atomic mass of these elements, as it was considered in [14-17]. Indeed, for Mo atoms the atomic radius  $R_{Mo}=140$  pm as well as the atomic mass  $M_{Mo}=96$  u are greater than the corresponding values  $R_{Fe}=127$  pm and  $M_{Fe}=56$  u of iron atoms (see Table I), so it is reasonable to expect a slowing down of diffusion processes caused by Mo. Let also notice that  $R_{Cr}=128$  pm and  $M_{Cr}=52$  u are comparable with these values for Fe which can explain the behavior of Cr atoms in the  $Fe_{78}Si_8B_{14}$  amorphous alloy. The case of Al is slightly different – on one hand the atomic radius of Al -  $R_{Al}=143$  pm is greater than  $R_{Fe}$  but on the other hand the atomic mass  $M_{Al}=27$  u is much smaller than  $M_{Fe}$ . It seems that these two factors play against each other and finally Al atoms do not influence the diffusion processes in the examined alloys.

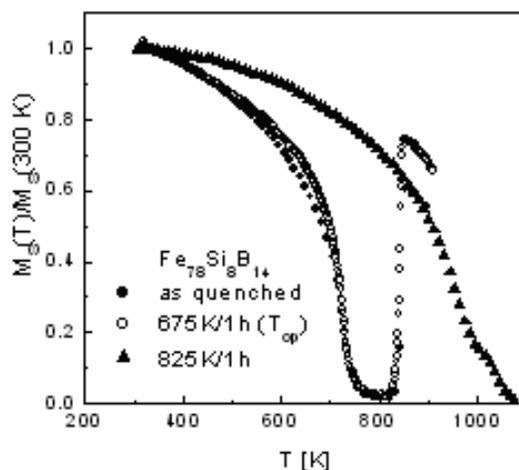


Fig. 9. Normalized magnetization versus temperature  $T$  for  $Fe_{78}Si_8B_{14}$  alloy (heating rate 5 K/min)

In order to study the mechanism of the ESMP effect magnetization in saturation was measured versus temperature for samples in the as quenched state and preliminary annealed at different temperatures. Fig. 9 shows three normalized curves  $M_S(T)$  determined for the  $Fe_{78}Si_8B_{14}$  alloy – i) in the as quenched state, ii) after optimization annealing at 675 K for 1 h and iii) after annealing at 825 K for 1 h. The temperatures  $T_C$  and  $T_X$  do not depend on thermal annealing up to the optimization annealing. For sample annealed at temperature  $T_a=825$  K the paramagnetic phase, characteristic for as quenched and optimized samples, is not observed. This means that during the preliminary annealing a partial crystallization (nanocrystallization) was already occurred. From the results presented in and Fig. 9 it is clear that the ESMP effect, observed in Figs. 3-6 cannot be attributed to formation of  $\alpha Fe$  or  $\alpha Fe(Si)$  nanograins. If the case the ESMP should be attributed to annealing out of free volume and internal stresses leading to formation of the so-called relaxed amorphous phase free of iron nanograins. It is proper to add that according to [9-11]

the relaxed amorphous phase contain small iron clusters with different magnetic order. Such a microstructure averages out magnetic anisotropy and gives the ESMP effect. Formation of the relaxed amorphous phase is a characteristic feature of the examined alloys which is very interesting from applications point of view because this phase ensures that the optimized alloys are not as brittle as the corresponding nanocrystalline alloys.

The main conclusions of the present paper can be summarized as follows:

- Alloying additions in the examined alloys cause a decrease of the Curie temperature  $T_C$ , an increase of magnetic permeability  $\mu$  and magnetization in saturation  $\mu_0 M_s$ .
- Magnetostriction coefficients:  $\lambda_{||}$ ,  $\lambda_{\perp}$  and  $\omega$  are sensitive on free volume content and decrease with the annealing temperature; in contrast to this saturation magnetostriction  $\lambda_S$  is independent on annealing temperature i.e. independent on free volume content.
- The ESMP (enhancement of soft magnetic properties) effect in the examined alloys can be attributed to the so-called relaxed amorphous phase free iron nanograins.

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