ROLE OF ADDITIONAL ELEMENTS ON CRYSTALLIZATION AND MAGNETIC PROPERTIES OF SOME NANOCRYSTALLINE Fe-Cu-Nb-Si-B ALLOYS[1]

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In $Fe_{75.5}Cu_1Nb_1Si_{11.5}B_9T_2$ and $Fe_{73.5}Cu_1Nb_1Si_{15.5}B_7T_2$ with T= V, Mo, W, Nb, and Ta, influence of the composition and additional alloying elements on controlled crystallization at 525°C and 550°C in Ar+H₂ was studied. The results obtained from ⁵⁷Fe Mössbauer spectra confirmed the influence of the chemical composition on the crystallization process. It was shown that the role of the additional alloying element, i.e. refractory metals partially replacing Nb, is strongly covered by changes in content of Si and B. With increasing diameters of the substituting element atoms the amount of the intergranular phase decreases.

Controlled crystallization of the amorphous alloys of the Fe-Si-B-Cu-(Nb,X) type yields a possibility to prepare nanocrystalline materials with excellent soft magnetic properties [1, 2, 3]. Changes of chemical composition influence temperature range of nanocrystalline state and/or magnetic properties. The content of iron and silicon determines the composition of nanocrystals formed [4] and thus some magnetic properties, e.g., saturation magnetization and magnetostriction. The content of boron should determine stability of the remaining amorphous phase. The additional alloying elements form nucleation centres of nanocrystals and/or influence the properties of the intergranular phase. Their atomic diameter could be an important parameter.

RELATIVE TRANSMISSION

1.00
0.97
0.94
0.90

AO(V)

AO(Nb)

BO(Nb)

BO(V)

-8   -4   0   4   8
VELOCITY [mm/s]
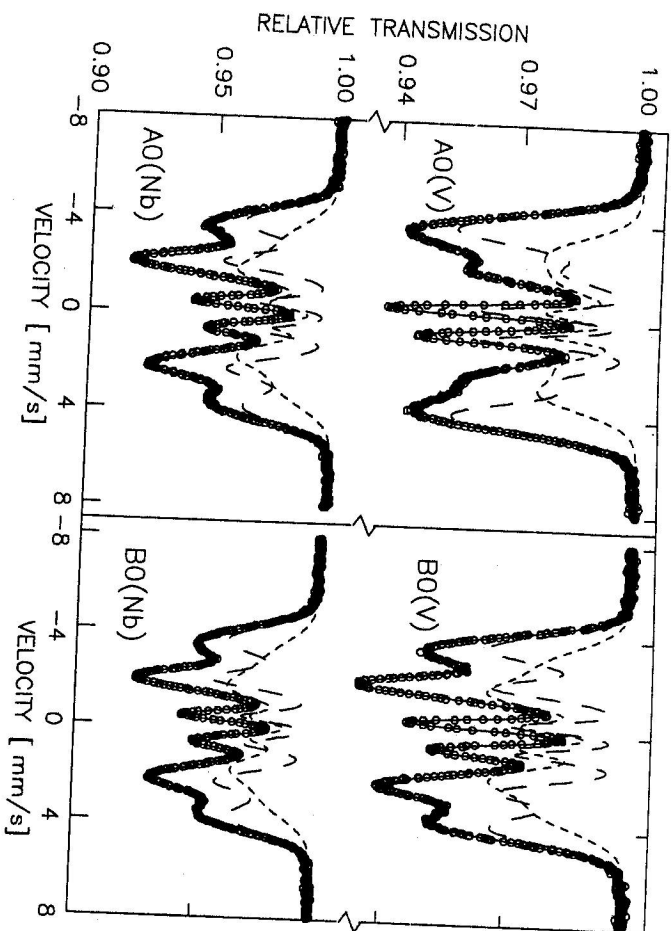
-8   -4   0   4   8
VELOCITY [mm/s]

Fig. 1. Mössbauer spectra of some amorphous samples. The dashed lines denote the two components described by two sextets with Gaussian distributions.

In this paper we have investigated the role of some additional alloying elements, refractory metals, on nanocrystalline state of two alloys that differ in Si and B contents. The refractory metals partially replace Nb. Recent x-ray and magnetic investigation of these alloys by Müller et al. [5] have shown that the substitution for Nb leads to the variation of the grain size of Fe-Si nanocrystals. The alloys with higher content of Si resulted in better soft magnetic properties probably because of their more suitable values of saturation magnetostriction and averaged magnetocrystalline anisotropy.

The ribbons of the amorphous alloys are of the same origin as in [5]. The nanocrystalline samples were prepared by annealing at 525°C and 550°C for 1 hour in Ar+H$_2$ atmosphere. Samples prepared from the alloy Fe$_{75.5}$Cu$_1$Nb$_3$Si$_{11.5}$B$_9$T$_2$ with T = V, Mo, W, Nb, and Ta were labelled AX(T), where X = 0, 1, 2 correspond to amorphous state, nanocrystalline state after annealing at 525°C, and nanocrystalline state after annealing at 550°C, respectively. The samples prepared from the alloy Fe$_{73.5}$Cu$_1$Nb$_3$Si$_{15.5}$B$_7$T$_2$ are denoted BX(T).

^{57}Fe Mössbauer spectra of the original amorphous and nanocrystalline states were taken at room temperature in transmission geometry. The spectra were deconvoluted in standard way supposing presence of crystalline and amorphous phases. This yielded intensities I of the components (taken as contents of phases) and their hyperfine pa-
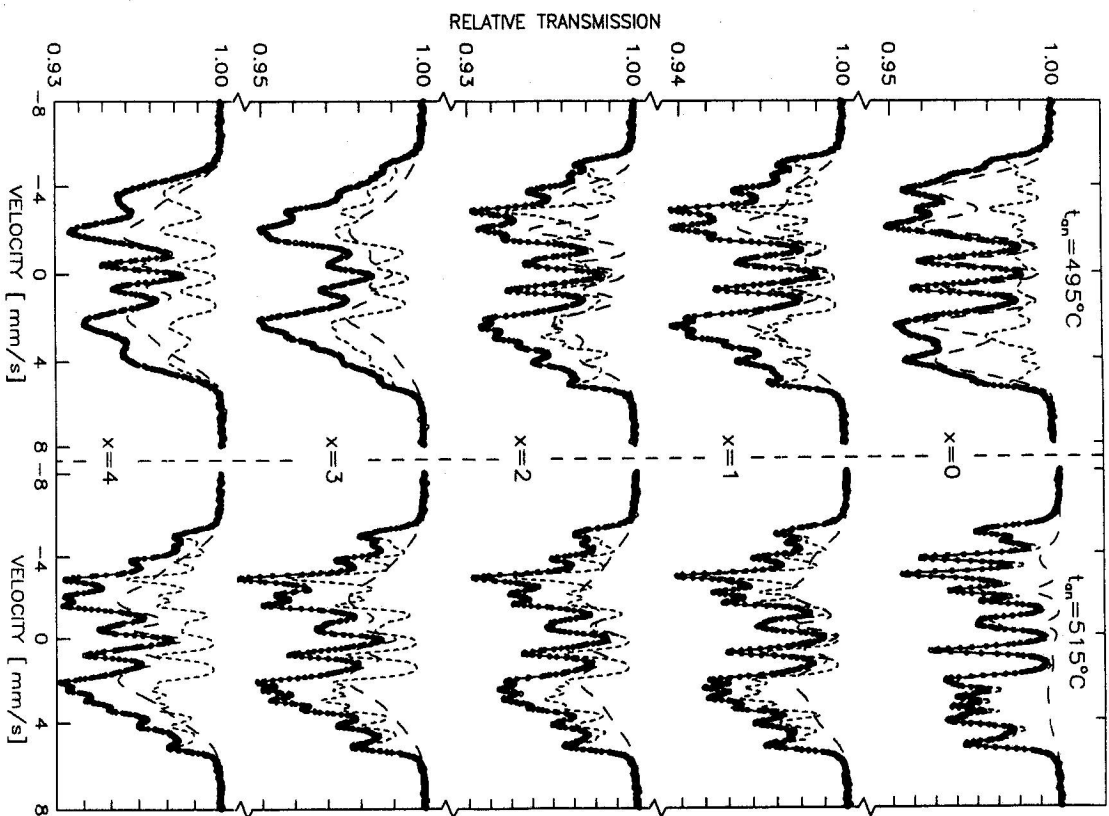
RELATIVE TRANSMISSION

1.00
0.95
0.93
1.00
0.95
1.00
0.94
0.93
1.00
0.95
1.00

t_{an} = 495°C

t_{an} = 515°C

x = 0

x = 1

x = 2

x = 3

x = 4

-8   -4   0   4   8
VELOCITY [mm/s]

-8   -4   0   4   8
VELOCITY [mm/s]

Fig. 2. Mössbauer spectra of the nanocrystalline samples after annealing at 550°C for 1 hour in Ar+H$_2$. The long-dashed lines denote the component ascribed to the intergranular phase and the short-dashed lines represent the crystalline part.

rameters: magnetic splitting B$_{hf}$ and isomer shift δ (given relative to α iron). Intensity ratio of the individual lines in sextets r, defined as ratio of the intensity of the second

content and decrease in Fe content.

In the nanocrystalline samples after annealing at 550°C only slight changes in comparison with previous nanocrystalline state can be observed as shown in Table 1. An increase in content in crystalline phases was observed in both series. The content of silicon in Fe-Si nanocrystals was estimated to 17 at.% and 20 at.% in A2 and B2 samples, respectively. It is in agreement with the increase and decrease in B_{hf}^c in A2 and B2 samples, respectively. Values of r in the A2 samples are slightly decreased in comparison with A1 samples, except the A2(Nb) sample. It seems that formation of a silicon poor Fe-Si surface layer in this alloy is connected with the state of the original amorphous ribbon. Pronounced changes of magnetization directions towards the sample plane normal simultaneously with the increase of δ of the intergranular phase occurred when annealing temperature was increased to 550°C in B2 samples. The changes in composition of the crystalline phase induced increase in Si and decrease in Fe concentration. This is in agreement with the observed increase of δ of the intergranular phase.

The amount of the crystalline phase displays a slight decrease with the increasing atomic diameter of the admixture (Fig. 3). The B_{hf} and δ are clearly determined by the content of Si and B. A simple dependence on the atomic diameter of the elements was not observed.

Comparison with macroscopic magnetic parameters published in [5] shows that there is no simple relation between B_s and B_{hf} derived here from the spectra. It could be due to different weight of individual contributions to B_{hf} and the differences in r value indicate that sufficiently high saturation field must be applied because of differences in magnetic structure between individual samples.

References

[1] G. Herzer and H. Warlimont: Nanostructured materials **1** (1992), 263;

[2] T. Zemčík, Y. Jirásková, K. Závěta, D. Eckert, J. Schneider, N. Mattern, R. Hesske: Materials Letters **10** (1991), 313;

[3] K. Závěta and Z. Kalva : in Nanomagnetism, ed. A. Hernado, 1993, Kluwer Academic Publishers, Dordrecht-Boston-London, p.207;

[4] G. Rixecker, P. Schaaf and U. Gonser: J. Phys.: Condens. Matter **4** (1992), 10295;

[5] M. Müller, N. Mattern, L. Illgen, H. R. Hilzinger and G. Herzer: Key Engineering Materials **81-83** (1993), 221;

[6] H.-D. Pfannes and H. Fischer: Appl. Phys. **13** (1977) 317;

[7] J. M. Grenecke, M. Henry and F. Varret: in Proc. ICAME, ed. V.G. Bhide, Jaipur, 1981, (Indian National Science Academy, New Delhi, 1982), p. 900;

[8] O. Schneeweiss, A. Sólyom, P. Marko, V. Frič: Hyperfine Int. **45** (1989), 249;

[9] O.K. Andersen, O. Jepsen nad D. Glötzel: Highlights of Condensed-Matter Theory (eds. F. Bassani, F. Fumi and M.P. Tosi, North-Holland, Amsterdam, 1985), p.59;

[10] O. Schneeweiss: J. Phys.: Condens. Matter **1** (1989), 4749;