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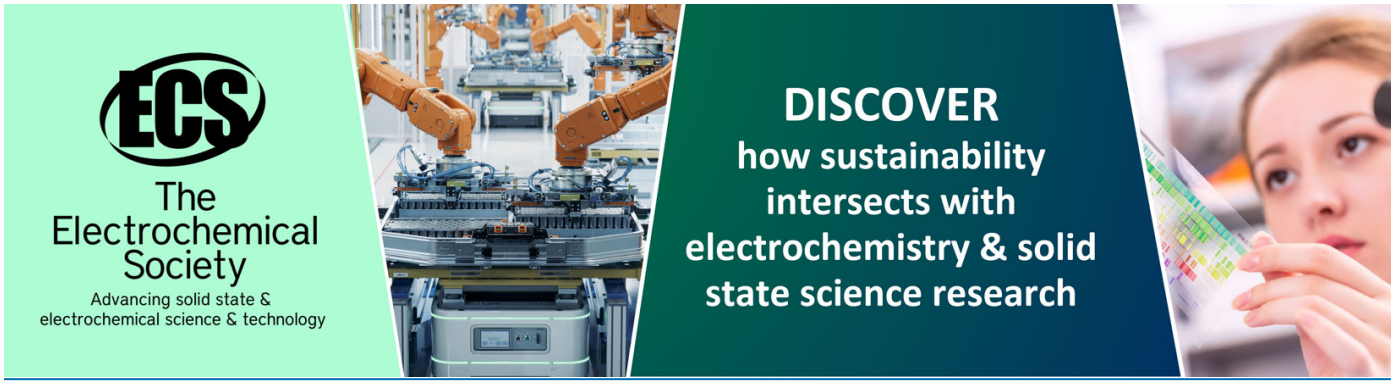

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Study of structural and magnetic properties of Co-substituted $(Fe_{100-x}Co_x)_{78}Si_9Nb_3B_9Cu_1$ alloys

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Abstract. Influence of controlled Co addition on structural and magnetic properties of $(Fe_{100-x}Co_x)_{78}Si_9Nb_3B_9Cu_1$ (x = 0, 20, 40, 60) alloys has been studied using DTA, XRD, magnetic measurements and Mössbauer spectroscopy. Results show that the variation of Co content affects the stability of the alloy against crystallization. Volume fraction of the nanograins varies only slightly and remains around 60 %. The corresponding grain diameter ranges between 13 to 15 nm. Obtained lattice parameter values suggest that the studied nanocrystalline samples consist of bcc Co-Fe phase with Si impurities, with Co content ranging between 38 to 61 % and Si content ranging between 7 to 15 %. Coercive field for as-cast specimens practically does not change with increase of Co content whereas for annealed samples it exhibits appreciable increases. Mössbauer measurements show that with increase of Co content, there is a change in: the environment around Fe-atom; spin texture and disorder in the studied specimens.

1. Introduction

Nanocrystalline materials of FINEMET type produced by controlled crystallization of amorphous ribbons [1] exhibit excellent soft magnetic properties making them attractive for technological applications. Ever since the discovery of FINEMET type alloys, attempts were made to further improve their soft magnetic behavior. Replacing Fe by Co seems to be quite effective for this purpose. Controlled Co addition [2-4] enhances the Curie temperature of the resulting nanocrystalline material while good soft magnetic properties are also observed at elevated temperature, thus making these materials appropriate for working at high temperature. It is worth noting that by reducing Si content

(Si content = 9) the Co is much more effective in inducing transversal anisotropy than for Si = 13 or 15 in Co substituted FINEMET-type alloys [5].

Table1. Lattice parameter (*a*), mean grain size (*D*) and volume fraction of nano-grains (V_x) for annealed specimens with different Co content (*x*).

x (%)	*a* (nm)	*D* (nm)	V_x (%)
0	0.2861 ± 0.0001	13.0 ± 1	62 ± 2
20	0.2848	14.5	60
40	0.2855	14.0	56
60	0.2855	15.0	60

Table2. Co content dependence of coercive field (H_c), specific magnetic moment (σ_S) and magnetostriction (λ_S) of as-cast and of annealed specimens.

x (%)	H_c (A/m)		σ_S (Am²/kg)		λ_S (ppm)	
	a.c.	ann.	a.c.	ann.	a.c.	ann.
0	20.7	22.9	146	157	15.4	4.1
20	19.9	46.3	147	151	11.5	9.8
40	20.8	48.4	137	131	11.4	11.1
60	18.3	91.3	123	135	11.9	13.3

In the present work we report the influence of controlled Co addition on structural and magnetic properties of $(Fe_{100-x}\ Co_x)_{78}\ Si_9Nb_3B_9Cu_1$ (x = 0, 20, 40, 60) alloys using differential thermal analysis (DTA), x-ray diffraction (XRD), magnetic measurements and Mössbauer spectroscopy.

2. Experimental

Ribbons of nominal composition $(Fe_{100-x}Co_x)_{78}Si_9Nb_3B_9Cu_1$ with x = 0, 20, 40 and 60 were prepared using a planar flow casting technique on copper wheel. The ribbons were about 20 µm thick and 4 mm wide. The as-cast (a. c.) samples were X-ray amorphous. Nanocrystallization in the studied samples was achieved by annealing them at 540 ^oC for1 h in the protective atmosphere of flowing Ar. First crystallization peak temperature (T_{X1}) was determined using DTA measurements performed at a heating rate of 20 K/min. XRD measurements were done using Cu-K_α radiation at room temperature and were analyzed by fitting crystalline components and amorphous components using pseudo-Voigt profiles to obtain lattice parameter (*a*), average grain size (*D*), and the volume fraction of the nanograins (V_x). Hysteresis loops were recorded at 28 Hz by using a fully digital hysteresis loop tracer based on a 12-bit oscilloscope. Specific magnetic moment (σ_S) was measured using a vibrating sample magnetometer with H_{max} = 700 kA/m. Magnetostriction constant (λ_s) was measured using Small Angle Magnetization Rotation method. Transmission Mössbauer spectra were recorded at room temperature, using ^{57}Co: Rh source. Mössbauer spectra were fitted with either distribution of hyperfine fields or overlapping of amorphous and crystalline components by using *NORMOS* program [5].

3. Results and discussions

T_{X1} of samples (DSC heating rate v = 20 K/min) with x = 0, 20, 40 and 60 are 458, 454, 461 and 447 ^0C respectively, showing that Co addition affects only slightly the stability of the alloy against crystallization. The error in Tx1 certainly depends on the inevitable variations in casting conditions as well, so better not to pay attention to these small variations in the crystallization temperature as a function of Co content.

Table 1 depicts the lattice parameter *a*, mean grain size *D* and volume fraction V_x of the nanograins with varying Co content. Perusal of table 1 shows that V_x remains almost unchanged (within experimental errors). Grain size of nanograins remains almost constant for the studied specimens, with lowest grain size value of 13.0 nm for sample with x = 0.

It is interesting to note that for the studied samples the volume fraction of DO_3 (Fe_3Si like) compound remains quite constant. Lattice parameter suggests that the studied nano- crystalline samples consist of bcc Co-Fe phase with Si impurities (Co content ranging between 38 to 61 % and Si content ranging between 7 to 15 %). The increase of Co content in the parent alloy produces the

increase of both Si and Co content in the nanocrystalline phase. Within experimental errors the first near neighbor distance in the residual amorphous matrix remains unchanged (0.249 nm ± 0.001) showing that the studied samples have similar mass density.

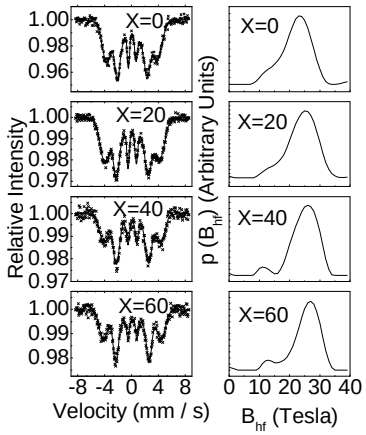

Figure1. Mössbauer spectra (experimental data and fit) and corresponding hyperfine field distributions for as-cast specimens with varying x.

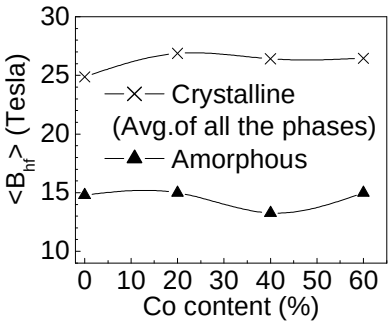

Figure2. Hyperfine field variation, $<B_{hf}>$, of the residual amorphous phase and of the crystalline phase (average $<B_{hf}>$ of all the crystalline components) as a function of Co content. Continuous lines are guide to the eye.

Table 2 depicts the coercive field (H_c), specific magnetic moment (σ_s) and magnetostriction (λ_s) for as-cast and annealed specimens with different Co content (x). Perusal of table 2 shows that H_c for as-cast specimens practically does not change with increase of Co content whereas for annealed samples it exhibits appreciable changes, where D also shows variation. Specific magnetic moment for amorphous and annealed samples decreases with increase in Co-content, which is in accordance with earlier results [2, 5]. Compositional dependence of λ_s in as-cast and annealed specimens is depicted in table 2. The highest λ_s value is displayed for the as-cast specimen having the lowest Co-content and decreases continuously with increase of Co-content, which is consistent with earlier reports [7]. After annealing, both H_c and λ_s increase with x. This can be due to internal stresses leading to inhomogenity in anisotropy. It should be noted that internal stresses does not necessarily disappear after nanocrystallization.

Mössbauer spectra and corresponding hyperfine field distributions for as-cast specimens with varying Co-content are shown in figure 1. Perusal of figure 1 shows that the hyperfine field distribution is broad and shows a low field component around 12 Tesla. This low field hump can be attributed to iron atoms having Nb atoms in their first near-neighbour shell, as was also observed in earlier studies [8]. This low field hump is more pronounced for higher Co-content, indicating that average numbers of Nb near neighbours to Fe are different in the specimens with higher Co-content.

Table 3 depicts the intensity of second and fifth lines relative to innermost lines of Mössbauer spectrum (b), average hyperfine field ($<B_{hf}>$), and width of hyperfine field distribution (ΔB_{hf}) for as-cast specimens with different Co content (x). Intensity of second and fifth lines relative to innermost lines of Mössbauer spectrum (b) is a measure of the spin texture in the specimen and can vary between 0 to 4 (completely random and spins within ribbon plane). Increase of b with increase in x, shows that more and more spins are getting aligned preferentially within ribbon plane. With increase of Co-content in the specimen, a continuous increase of $<B_{hf}>$ indicates the change of environment around Mössbauer (Fe) atom. $\Delta B_{hf} / <B_{hf}>$ is a measure of disorder in the specimen. $\Delta B_{hf} / <B_{hf}>$ decreases with increase of Co-content in the specimen, indicating decrease of disorder in the specimen.

Figure 2 shows the variation of $<B_{hf}>$ for both crystalline (average $<B_{hf}>$ of all the crystalline components) and residual amorphous matrix with varying Co content. Perusal of figure 2 shows that $<B_{hf}>$ for both crystalline and amorphous phases vary with different Co content, indicating differences

in the environments around Fe atom in the studied specimens. Low values of $<B_{hf}>$ observed for the residual amorphous matrix (as compared to nanocrystalline phase) are consistent with the presence of boron as near-neighbors of Fe [9]. The moderate values of H_c also support the conjecture that boron remains in the residual amorphous matrix.

Table 3: Intensity of second and fifth lines relative to innermost lines of the Mössbauer spectrum (*b*), Average hyperfine field ($<B_{hf}>$), width of hyperfine field distribution (ΔB_{hf}) for as-cast specimens with different Co content (*x*).

x	*b*	$<B_{hf}>$ *(T)*	$\Delta B_{hf} / <B_{hf}>$
0	2.31± 0.07	22.07± 0.33	0.227
20	2.26	23.85	0.217
40	3.39	24.56	0.199
60	3.34	24.88	0.200

Width (*W*) of the crystalline line of the Mössbauer spectrum for samples with x = 0, 20, 40 and 60 is respectively 0.489, 0.435, 0.426 and 0.362 mm/s. Decrease of *W* with increase of Co content in the specimen shows decrease of chemical disorder in the nanocrystalline specimens. As Co content in the alloy increases, the area of the crystalline components increases slightly (between 75 and 81 %) in accordance with the small variation of the crystalline volume fraction determined by XRD.

4. Conclusions

Variation of Co content affects the stability of the alloy against crystallization. Volume fraction of the nanograins varies slightly around 60 % and the corresponding grain diameter ranges between 13 to 15 nm. Lattice parameter values suggest that the studied nanocrystalline samples consist of bcc Co-Fe phase with Si impurities, with Co content ranging between 38 to 61 % and Si content ranging between 7 to 15 %. Coercive field for as-cast specimens practically does not change with increase of Co content whereas for annealed samples it exhibits appreciable changes. Mössbauer measurements show that in as-cast samples with increase of Co content, there is a difference of average number of Nb near-neighbors to Fe, more spins are aligned preferentially within ribbon plane. Hyperfine field values of the residual amorphous matrix (within nanocrystalline specimens) suggest that boron is retained in the residual amorphous matrix. Observed moderate values of coercive field also support this conjecture.

Acknowledgements

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