Structure of Fe-Cu-Nb-Si-B Alloys with Various Copper Concentration

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Abstract. In this paper, for alloys: Fe\textsubscript{0.2}Cu\textsubscript{0.3}Nb\textsubscript{3}Si\textsubscript{16.5}B\textsubscript{6}, Fe\textsubscript{0.1}Cu\textsubscript{1}Nb\textsubscript{3}Si\textsubscript{16}B\textsubscript{6}, Fe\textsubscript{0.15}Cu\textsubscript{1.5}Nb\textsubscript{3}Si\textsubscript{16.5}B\textsubscript{6} inhomogeneity of structure in thickness is shown with using scanning and transmission electron microscopy, crystalline phases are determined. It is noted that a multiphase magnetic structure is observed in an alloy with a low copper content upon complete crystallization.

1. Introduction

As is known, some properties of solids are structurally sensitive, which is also true for amorphous materials. For example, Fe-Si-B-Cu-Nb semi-crystalline alloys are widely used in industry. Properties of such alloys obtained by rapid quenching method combine most attractive characteristics of both amorphous and crystalline structures. Knowledge of structure, as well as mechanism of its formation gives, understanding of known amorphous materials properties. And secondly it will allow them to be improved, to obtain materials with specified characteristics.

2. Objects and research methods

Three alloys: Fe\textsubscript{0.2}Cu\textsubscript{0.3}Nb\textsubscript{3}Si\textsubscript{16.5}B\textsubscript{6}, Fe\textsubscript{0.1}Cu\textsubscript{1}Nb\textsubscript{3}Si\textsubscript{16}B\textsubscript{6}, Fe\textsubscript{0.15}Cu\textsubscript{1.5}Nb\textsubscript{3}Si\textsubscript{16.5}B\textsubscript{6} were investigated by X-ray diffraction on Bruker D8 Advance. Samples were prepared for scanning electron microscopy (Carl Zeiss Ultra+) and transmission electron microscopy (Carl Zeiss Libra 200). Also has been carried out study of alloys elemental composition.

3. Results and discussion

According X-ray diffraction profiles shown in Figure 1 [1], all samples are characterized by presence of two amorphous halos. Both amorphous halos are located at angles $\theta_1 = 45^\circ$ and $\theta_2 = 80^\circ$. For the Fe\textsubscript{0.15}Cu\textsubscript{1.5}Nb\textsubscript{3}Si\textsubscript{16.5}B\textsubscript{6} alloy, an additional crystalline peak is observed at the angle $\theta = 66^\circ$. All peaks observed on X-ray diffraction patterns correspond to the $\alpha$-Fe phase. It seemed that in this way we can talk about X-ray amorphousness of these alloys, with a small amount of crystalline phase at 1.5\% copper content. However, when examining amorphous matrix by thickness, electron microscopic images of all foils contain some spherical inclusions, like those of the Fe\textsubscript{0.15}Cu\textsubscript{1.5}Nb\textsubscript{3}Si\textsubscript{16.5}B\textsubscript{6}, sample, Figure 2, having various sizes, from tens nanometers by two microns. Their location along thickness of samples are random, however, more of them were recorded close to contact surface of the alloy (for Figure 2, this is lower edge on image). It was not possible to establish an explicit dependence of quantity and their size on percentage of copper in alloy. In some SEM images, only “footprints” of inclusions in amorphous matrix were found.
Figure 1. X-ray profiles obtained with the free side [1]

Figure 2. SEM images of the ends in the initial state.

Figure 3 shows an example of X-ray profiles taken from both sides of one alloy. Crystalline peaks are present in X-ray diffraction patterns from contact surface, which are absent in profiles obtained from free side. Moreover, these peaks are at same angles as crystalline peak in X-ray diffraction
patterns obtained from free side for FeCu$_{1.5}$Nb$_3$Si$_{16.5}$B$_6$ sample. Profiles of X-ray diffraction patterns shown in Figure 3 and electron microscopic images in Figure 2 suggests that considered alloys are characterized by structure stratification in thickness [2]. Crystalline phase is formed mainly at contact side of samples, and type of this phase is same for all alloys. By the way more copper in alloy composition, greater fraction of the crystalline phase in it, which correlates with literature data. [3].

Figure 3. X-ray diffraction patterns of FeCu$_{0.2}$Nb$_3$Si$_{16.5}$B$_6$ from two sides.

Figure 4. Results of measuring elements concentration along the line by thickness of sample FeCu$_{0.2}$Nb$_3$Si$_{16.5}$B$_6$

Also, a qualitative and quantitative elemental analysis was carried out to clarify how distribution of elements over alloys thickness. As seen in Figure 4, the elemental composition of the amorphous matrix is isotropic in thickness. This is typical for all considered alloys; the elemental composition of
spherical inclusions was also measured, however, no noticeable changes relative to amorphous matrix were recorded.

Bright-field images of two alloys (FeCu_{0.2}Nb_{3}Si_{16.5}B_{6}, FeCu_{1.5}Nb_{3}Si_{16.5}B_{6}) were obtained at several magnifications, with different degrees of defocusing Figure 5.

Contrast analysis of transmission electron microscopic (TEM) images identified dark spots in both samples as nanocrystals about 4-8 nanometers in size (highlighted by arrows in Figure 5). To clarify the type of phase in nanocrystals, electron diffraction patterns were obtained.

Figure 5. Bright-field TEM images of FeCu_{0.2}Nb_{3}Si_{16.5}B_{6} with different defocusing degrees.

Figure 6. TEM image of FeCu_{1.5}Nb_{3}Si_{16.5}B_{6}, sample, with electron diffraction pattern and profile.

Figure 7. TEM image of FeCu_{0.2}Nb_{3}Si_{16.5}B_{6}, sample, with electron diffraction pattern and profile.
Figures 6 and 7 show electron microscopic images of FeCu$_{0.2}$Nb$_3$Si$_{16.5}$B$_6$, FeCu$_{1.5}$Nb$_3$Si$_{16.5}$B$_6$ samples, from which diffraction patterns were obtained, and corresponding electron diffraction patterns. The result decoding of electron diffraction patterns is presented in Table 1: the first column is number of identified ring; 1/r$_{exp}$ — radius of ring in reciprocal space in reverse angstroms; r$_{exp}$ — radius of ring in direct space in angstroms. The fourth column shows theoretical value 1/r$_{theor}$ of corresponding phase with its decoding.

| №  | FeCu$_{0.2}$Nb$_3$Si$_{16.5}$B$_6$ | FeCu$_{1.5}$Nb$_3$Si$_{16.5}$B$_6$ |
|----|---------------------------------|---------------------------------|
| 7  | 1/r$_{exp}$; 1/A | 1/r$_{theor}$; 1/A | r$_{exp}$; A | r$_{exp}$; A | 1/r$_{theor}$; 1/A |
| 6  | 9,556 | 1,05 | 9,562 FeSi$_2$ | 8,043 | 1,24 | 8,044 FeSi$_2$ |
| 5  | 8,228 | 1,22 | 8,203 FeB | 6,678 | 1,5 | 6,673 FeB |
| 2  | 6,752 | 1,48 | 6,743 FeB | 4,76 | 2,1 | - |
| 1  | 4,907 | 2,04 | 4,906 FeB | 3,82 | 2,6 | - |

4. Summary
In this paper, was shown that in alloys with Fe-Si-B-Cu-Nb type with copper content from 0.2 to 1.5 at.%, anisotropy of structure along thickness was observed. All alloys are amorphous-crystalline, crystal size ranges from 5 nm to 2 microns and has a complex phase composition of iron borides and iron silicides. In this case, elemental composition is uniform in amorphous phase and when comparing the amorphous structure and crystalline phase.

5. References
[1] Tkachev V V, Tsesarskaya A K, Ilin N V, Krainova G S, Fedorets A N, Plotnikov V S, Poliansky D A 2017 The microstructure and magnetic properties of the Fe-Cu-Nb-Si-B finemets with different copper content AIP Conference Proceedings 1874 art 040051
[2] Silveyra J M, Vlasak G, Svec P, Janickovic D, Cremaschi V J 2010 Domain imaging in FINEMET ribbons Journal of Magnetism and Magnetic Materials Vol 322 18 2797-2800
[3] Wojciechowski K 2005 Monte Carlo Simulations of Model Particles Forming Phases of Negative Poisson Ratio Properties and Applications of Nanocrystalline Alloys from Amorphous Precursors Vol 184 241-252

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