PHASE TRANSFORMATION IN Fe-B-(Si)-C-(Cu) SOFT MAGNETIC SYSTEMS

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1. Introduction

Nanocrystalline alloys based on eutectic Fe-B are in general very interesting mainly for their unique magnetic properties. The bcc-Fe small grains (size less then 100nm) immersed in amorphous matrix exhibit low saturation magnetic flux density (B_s), high permeability (μ_e) and low coercive force (H_c) as well as magnetostriction close to zero (λ) [1-5]. Adding of Si can next improve its magnetic properties making this nanostructured material suitable for distributions transformers and/or sensors [6-8]. Tuning the structure by adding a small amount of Cu as a nucleating agent can next lead to the improved formation of large number of small clusters from amorphous matrix, so that creating a greater number of smaller crystalline bcc-Fe grains [6, 9, 11]. The Fe-B-Si-P systems in combination with a small amount of C where P and C were added at the expense of B also have been frequently studied [8, 10-12]. This combination of alloying elements into the eutectic Fe-B system can lead to the improved glass forming ability and then resulting physical properties, which are dependent on the final crystalline structure and its proportion to the amorphous matrix. Only a few added combination of B-Si-C have been studied so far [9]. To cover this gap in our work the $(Fe_{85}B_{15-x}Si_x)_{98-y}C_2Cu_y$ systems, where x=0; 5 at.%, and y= 0; 1 at.% were investigated. The aim was to achieve similar (or better) properties using lower number of inexpensive elements as by adding the more expensive or rare ones (Co, Ni, Mo, Pd). We investigated the phase evolution by controlled structural transformation using isothermal or linear heating. The dependence of the onset of crystallization - T_x of transformed ferromagnetic phase bcc-Fe on different thermal treatment was analyzed.

2. Experimental procedure

Amorphous ribbons 6 mm wide and ~20 μm thick were prepared by the planar flow casting (PFC) technique. The samples were linearly heated with 10K/min heating rate and also isothermally annealed at several selected temperatures; the sequence of crystallization stages of the amorphous structure was thus observed. Dependencies of the electric resistivity on time and temperature were monitored in terms of relative electric resistivity (standardized ratio $R(T)/R(T_0$=300K). This method enables us to define the beginning of the crystallization in the system quite exactly. Structure of the as-cast samples as well as of isothermally annealed samples was investigated by X-ray diffraction (XRD) using Bruker D8 diffractometers and by transmission electron microscopy (TEM) using JEOL 2000FX at 200 kV. The structure and phase evolution of bcc-Fe (at the expense of the matrix in the Fe-B-C and/or Fe-B-Si-C based

systems) was also analyzed by in-situ TEM observation during isothermal annealing of samples at 713K and 663K.

3. Results and Discussion

The amorphous state of as-quenched samples was checked by X-ray diffraction. The patterns from ribbons in the as-cast state exhibit only a typically broad hallo, Fig. 1. After the isothermal annealing at 633K/30min the samples exhibit growing amount of a bcc-Fe phase. Its content in the alloys after regulated crystallization is shown in Fig. 1, too. The annealing temperature was chosen according to the resistivity measurements, shown in Fig. 2a. By comparing the diffraction patterns and resistivity behavior it is evident that the addition of Si shifts the crystallization onset and also changes the kinetics of crystallization process (shape of the resistivity decrease, Fig. 2a).

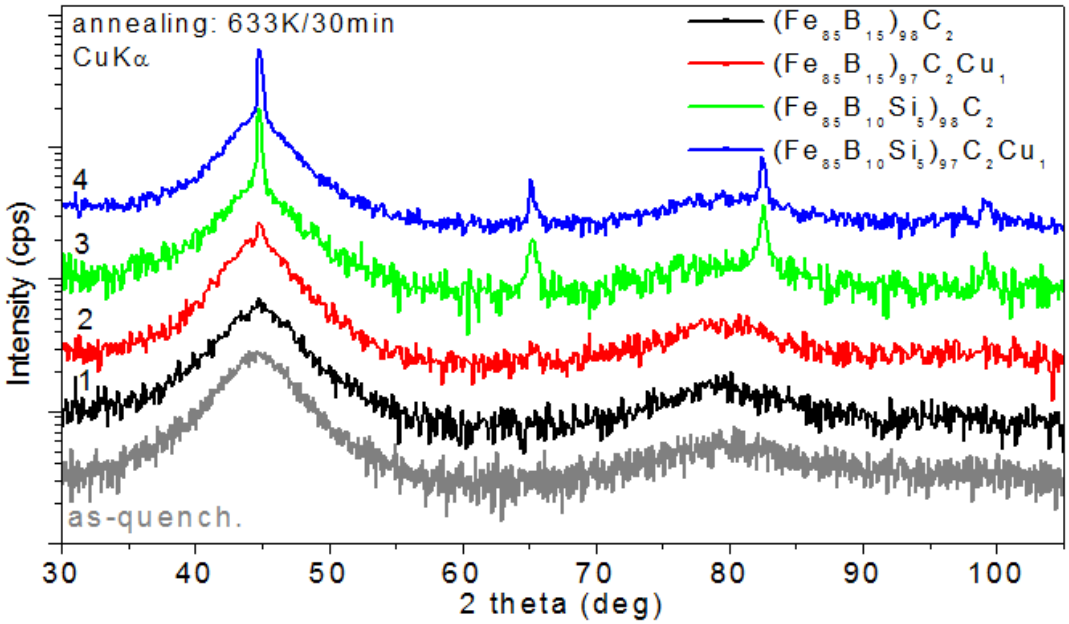

Fig. 1 *XRD patterns taken from the marked samples in as casts state and after isothermal annealing on 633K for 30min (curve 1 - $(Fe_{85}B_{15})_{98}C_2$; curve 2 - $(Fe_{85}B_{15})_{97}C_2Cu_1$; curve 3 - $(Fe_{85}B_{10}Si_5)_{98}C_2$; curve 4 - $(Fe_{85}B_{10}Si_5)_{97}C_2Cu_1$).*

This transformation takes place typically in more stages. In the first stage the crystallization of ferromagnetic phases takes place, followed by the evolution of borides phases in subsequent stages. The crystallizations onsets are visible in Fig. 2a. as smooth decrease of electrical resistivity. Shape of the transformation curves reflects the nature and the kinetics of these phase changes. The Fe-B-C alloy is an exception to this behavior as it exhibits only a single stage transformation at higher temperatures. This effect is similar to that observed in Fe-B-P system [11, 12]. Similar effect of additions to basic Fe-B-P/Fe-B-C systems appears with addition of Cu, which influences the temperatures of crystallizations onsets.

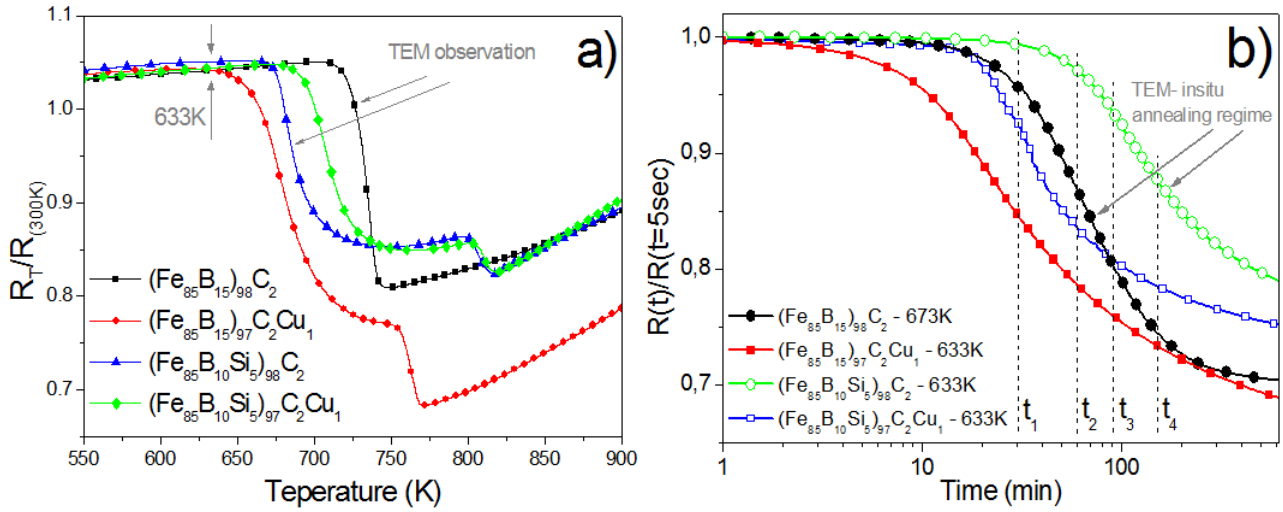

Fig. 2: a) *Temperature dependence of the relative electrical resistivity for the systems with different chemical composition, marked - the temperature chosen for isothermal annealing for XRD analysis;* b) *Evolution of relative electrical resistivity for the studied systems during isothermal annealing at 633K and 673K (t_1=30min, t_2=60min, t_3=90min, t_4=150min).*

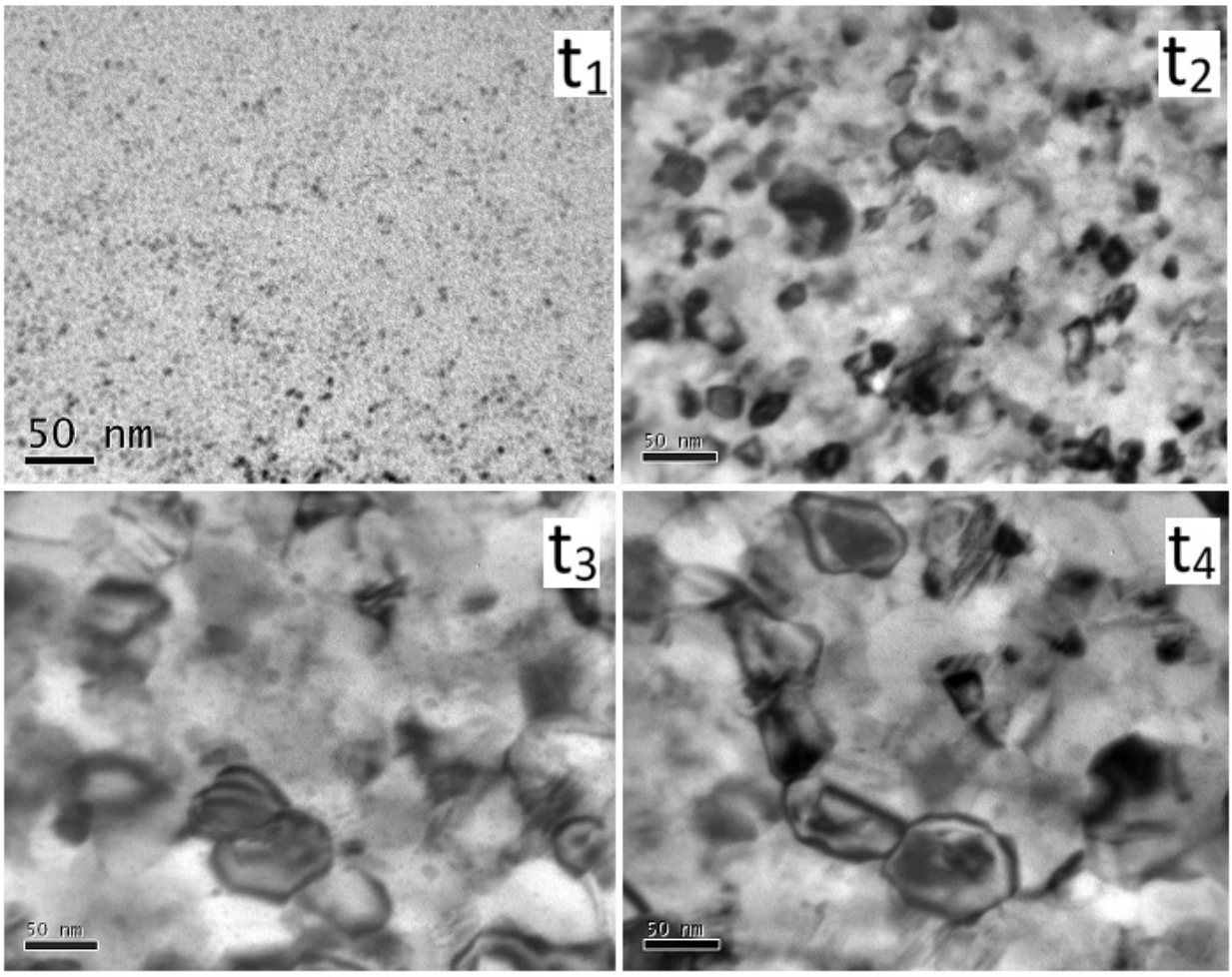

Fig. 3 *Structure evolution by TEM image of rapidly quenched $(Fe_{85}B_{10}Si_5)_{98}C_2$ system observed during in situ isothermal annealing in time: t_1, t_2, t_3, t_4.*

Kinetics of structural changes has been observed also by resistivity evolution during isothermal annealing, shown in Fig. 2b for 633K. For the Fe-B-C sample the higher annealing temperature – 673K – is shown, because this sample exhibits higher temperature of crystallization onset. Thermal regime for the TEM observation by in-situ annealing for the sample Fe-Si-B-C (Fig. 3) and Fe-B-C (Fig. 4) has been selected on the basis of the time evolution of crystallization from Fig. 2b. The aim of this experiment was to monitor the evolution of grain size and morphology with proceeding transformation and the differences due to the addition of Si into the basic Fe-B-C system. The growth of crystalline phase at the expense of the amorphous matrix is illustrated in Fig. 4. By comparison of the resulting structures the effect of Si content is evident: the grains of bcc-Fe are smaller and more regular.

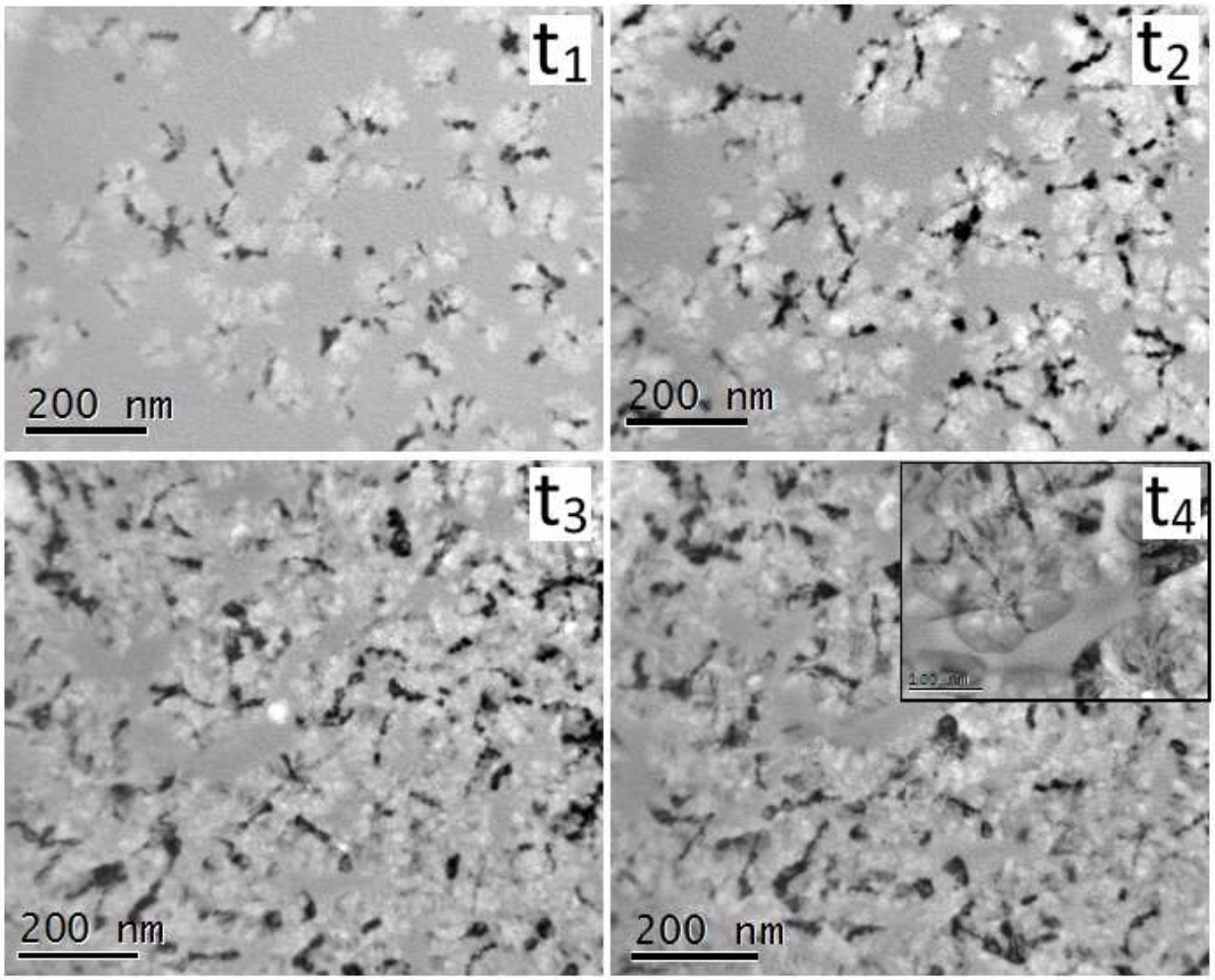

Fig. 4 *Structure evolution by TEM image of rapidly quenched* $(Fe_{85}B_{15})_{98}C_2$ *system observed during in situ isothermal annealing:* t_1, t_2, t_3, t_4.

4. Conclusion

The study of transformation of the systems based on Fe-B-(Si)-C-(Cu) indicated the influence of individual addition or the combination of additions of Si and Cu into the basic Fe-B-C system on grain size morphology and final structure. This complex metastable system transforms in two stages, as proven by resistivity changes. There is the exception of a simple Fe-B-C system where a single stage transformation occurs. This can serve as a basis for comparison with

similar system behavior of Fe-B-P [11, 12]. The phase analysis reveals the content of ferromagnetic bcc-Fe phase in different contents for various compositions obtained by the same thermal processing. The content of Si leads to the smaller and more regular bcc-Fe grains, which should guarantee to achieve improved magnetic properties (with low anisotropy) and increase of temperature interval between the two crystallization stages, resulting in a better stability of the ferromagnetic phase, as desired. The addition of Cu makes a small shift of crystallization onset temperature. The used combination of additions in sample Fe-B-Si-C-Cu has lead to the required stability increase. Therefore, it makes this composition perspective for use in practical applications and so will be the subject for further more detailed studies.

Acknowledgement

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