PHASE TRANSFORMATIONS IN ALPHA-IRON WITH BERYLLIUM COATINGS AFTER ISOCHRONOUS AND ISOTHERMAL ANNEALING

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1. INTRODUCTION

The α-Fe foils with the beryllium coatings, produced by magnetron sputtering, is studied here. This system is considered as a good candidate for thermonuclear application. The phase formation processes in the courses of isochronous and isothermal annealing are studied by means of three registration methods. The Mössbauer spectroscopy is performed by the back-scattering geometry technique based on conversion electrons (CEMS) and the absorption geometry technique based on γ-rays. X - ray diffraction is used for confirmation of Mössbauer investigation results. The Rutherford back scattering method is applied for studying the Fe distribution over the specimen depth in the course of annealing. In order to observe the initial stage of phase formation in the diffusion zone by CEMS at a low annealing temperature, the Be coating of a thickness about 1μm is used. The thickness of the Fe specimen is about 10 μm. The chosen Be concentration doesn't exceed the limit of solution in Fe within the range of annealing temperature in use (500 to 1000°C).

2. EXPERIMENTAL

The specimens have been made of the α-Fe foils (enriched by ^{57}Fe isotope up to 89 at. %), rolled to the thickness 10-13 μm and subject to homogenization annealing in vacuum at 800°C for 2 hours. The deposition of Be onto the iron foil have been carried out by magnetron sputtering at the original facility "Argamak" [1]. The beryllium coatings 1.8 μm and 0.8 μm thick for isochronal annealing and 0.5μm for isothermal one are used. Half-an-hour isochronal annealing series have been carried out within the temperature range from 500 to 1000°C with the 50°C interval. Isothermal annealing series have been carried out at two temperatures - 675 and 720°C. The annealing vacuum was of $5 \cdot 10^{-6}$ mm Hg.

The Mössbauer studies have been performed at a room temperature by means of two registration techniques: the technique of conversion electrons Mössbauer spectroscopy (CEMS), from both sides of a specimen in back scattering geometry and the γ-ray technique in absorption geometry. In the first case, information is obtained about a phase state of subsurface layers with the thickness values that depend on their chemical composition. For the iron layer the maximum thickness comprises ~400 nm [2]. The CEMS studies of the specimen

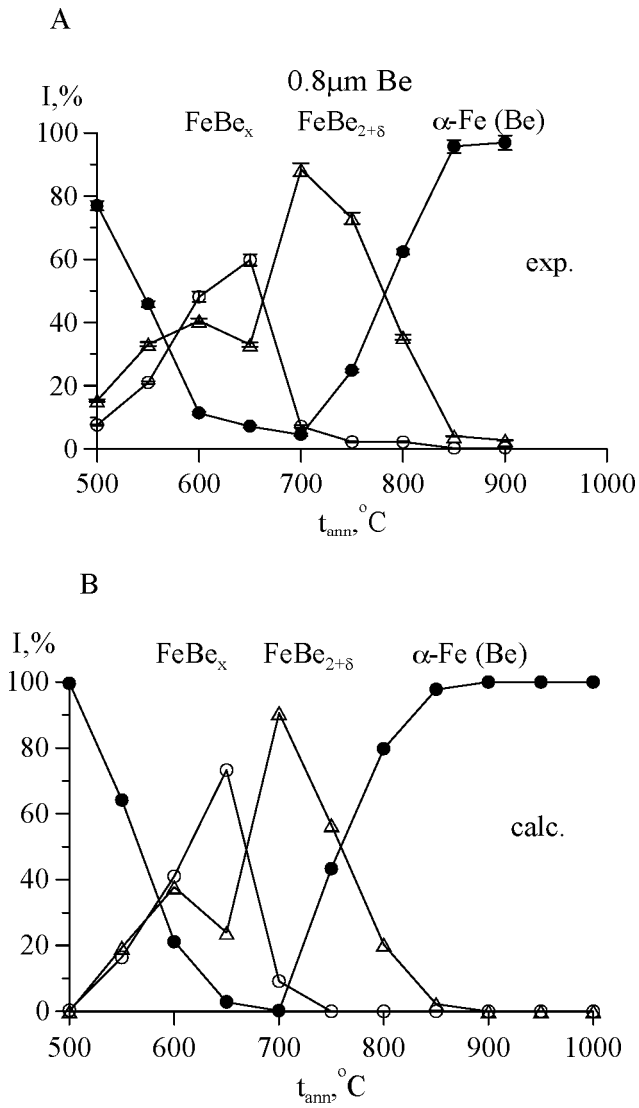

Fig 1. The experimental (A) and calculated (B) relative intensities of the partial CEMS spectra of ^{57}Fe nuclei in the binary system Fe(13μm)-Be(0.8μm) versus the temperature of subsequent isochronous annealing.

back side are related to possible changes in a phase state in the sample large depths in the course of annealing. With the second technique, Mössbauer spectroscopy provides the phase-state information averaged over the specimen thickness. The method of restoration of the hyperfine parameter partial spectrum distribution function [3] has been used for analyzing the Mössbauer spectra. The method is realized in the DISTRI computer code of the package *MSTools* [4].

Control for a deposited Be layer depth as well as determination of the iron concentration profile over the depth have been implemented by the Rutherford back scattering method (RBS) at the electrostatic accelerator UKP-2-1 (the Kazakhstan Institute of Nuclear Physics) [5].

3. RESULTS AND ANALYSIS

Isochronous annealing

The Be-Fe binary system incorporates several intermetallic compounds: $FeBe_2$, $FeBe_5$ and $FeBe_x$ [6-8], in addition to regions of solid solution of beryllium in iron (up to 21 at.% at 860°C) and iron in beryllium (up to 0.2 at.% at 850°C).

The compound $FeBe_2$, or the β-phase, being ferromagnetic, has the *hcp* (hexagonal close packed) crystalline structure of the $MgZn_2$ type. A wide range of homogeneity from 67 up to 79 at. % Be is typical for this phase. Due to this, below we'll denote it as $FeBe_{2+\delta}$ ($0 \le \delta \le 1.8$). When δ increases, the Curie point of the β phase decreases from 611 K (at $\delta=0$) to ~300 K (at $\delta \sim 1.8$). Therefore, this phase is in magnetically ordered state at room temperature, practically, through the entire concentration range. The $FeBe_5$ compound, or the ε-phase, possesses the UNi_5-type cubic crystalline structure. $FeBe_5$ has the $MgCu_5$-type structure referring to the spacial group Fd3m. This phase is in paramagnetic state at a room temperature. Beryllides with the highest contents of beryllium, obeying a general formula $FeBe_x$ ($11.2 \le x \le 11.8$) and being paramagnetic, possess hexagonal structure. Mössbauer spectra of these compounds at a room temperature are lines of paramagnetic types [9].

The identification of partial spectra has been carried out in accordance with the data on the beryllium compound Mössbauer spectra. The partial spectrum of paramagnetic type with a value of quadruple displacement ε ranging from 0 up to 0.4 mm/s has been assigned to the ε-phase $FeBe_5$ and to the compounds with higher content of beryllium (this partial spectrum is denoted here as $FeBe_x$). The partial spectrum with the set of the magnetic field values (H_n) ranging from 175 to 195 kG has been assigned to the β-phase $FeBe_{2+\delta}$, and the spectrum with the range of the magnetic field from 260 to 340 are associated with solid solutions of Be in the α-iron, denoted as α-Fe(Be).

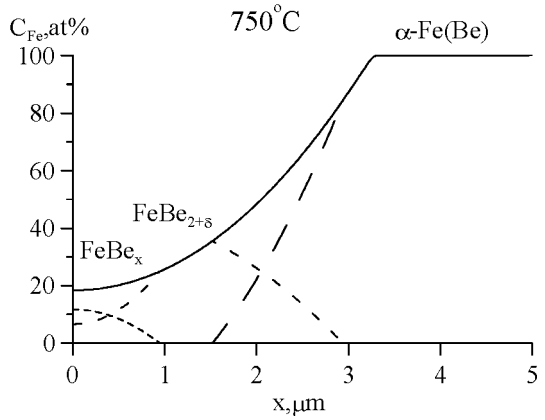

Fig. 2. The calculated distribution of Fe atoms referring to various phases, over the depth for the system Fe(13μm)-Be(1.8μm) at temperature of 750°C of subsequent isochronous annealing.

As an example, relative intensities of the partial CEMS spectra taken from the Be deposited side versus various annealing temperatures (t_{ann}) are shown in Fig.1A. The coating thickness is 0.8 μm. It should be noted that isochronous annealing series, lasting 30 min each, are carried out within the temperature range from 500 to 1000°C with the 50°C interval.

The studied annealing temperature range (500÷1000°C) may be conventionally divided into three regions: law temperatures (t_{ann}≤600°C), where the beryllium-contained phases are nucleating; the middle region (600°C<t_{ann}<750÷850°C), where the forming beryllium-contained phases are comparable, and the high-temperature region (t_{ann}>750÷850 °C), where disintegration of formed phases and diffusion of Be atoms into the α-Fe(Be) solution take place., being quite clear, provided one takes into account an additional inflow of the surface Be atoms to the diffusion zone.

The physical model of diffusion and phase formation in the two-layer binary Fe-Be system has been proposed. The model takes into account the mutual diffusion mechanism, peculiarities of phase diagram Fe-Be. The computer code that implements the proposed model has been developed. The code describes the phase formation kinetics in any region of the two-layer binary system at isochronous annealing (see Fig.1B and Fig.2).

Good agreement between the calculations and the Mössbauer investigations of both the interior and the subsurface layers of the two-layer binary system Fe-Be has been obtained. It means that frequency factors and the energies of activation of iron in beryllium and beryllium in iron are in good agreement with appropriate literature data. The diffusion coefficients don't depend on a formed phase state in specimen depth.

The distribution of Fe atoms, referring to various phases, over the depth of the system Fe(13μm)-Be(1.8μm) has been calculated for various temperatures of subsequent isochronous annealing (see for example Fig.2).

Isothermal annealing

The first half-an-hour annealing of α-iron foils 11 μm thick with the berylliym coating 0.55μm thick at temperatures 675 and 720°C, leads to appearances in the sample subsurface layers from deposited side mainly $FeBe_2$ with the small amount of solid solution of Fe(Be). The distribution of phase relative intensities in subsurface layers and in the bulk after first half-an-hour annealing depend on thickness of Be coating, time and velocity of temperature increasing and annealing temperature.

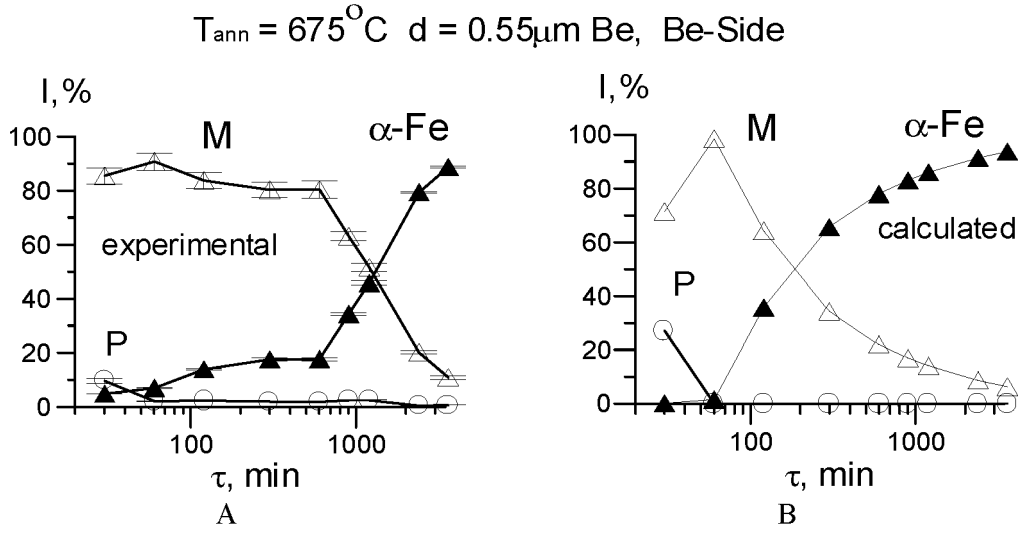

Fig.3 The experimental (A) and calculated (B) relative intensities of the partial CEMS-spectra in system Fe(11μm)-Be(0.55μm) versus the time of isothermal annealing at temperature of 675°C.

The following time increasing of isothermal annealing leads to disintegration of $FeBe_2$ compound on solid solution. In the case of isothermal annealing at temperatures 720°C this process is more fast than the same one at temperature 670°C.

As it should be expected, phases with the lower beryllium content are disposed deeper from the surface . As the time of subsequent isothermal annealing increases, the solid solution α-Fe (Be) is getting closer to a specimen surface and diffusion process is completed after formation along sample depth of equal composition solid solution. Qualitatively this tendency is kept and for absorption geometry of Mössbauer spectroscopy and for X-ray diffraction (see Fig.3-5). According to evaluates X-ray diffraction method in our case receives information from depth up to 8 μm.

To study kinetics of isothermal annealing more detail we use mentioned above Computer code, developed on the base of Darken theory of mutual diffusion. All model parameters and conditions which have been used for description of isochronous annealing are used again and for isothermal ones. The temperature dependence of diffusion coefficients for isothermal process is used according to Arrenius law. In calculation the following parameters are used: for the diffusion coefficient of iron in beryllium D_{10}=1 cm^2/s and Q_{10}=220 kJ/mole [12,13], whereas for the diffusion coefficient of beryllium in iron D_{20}=0.1 cm^2/s and Q_{20}=241.2 kJ/mole [14]. The frequency factor D_{i0} and activation energy Q_i have been proposed in literature for high temperature range, nevertheless they are appropriate for annealing at t_{ann} = 720°C (see Fig.4B). But for isothermal annealing at t_{ann} = 675°C these diffusion coefficients too large (see Fig.3B).

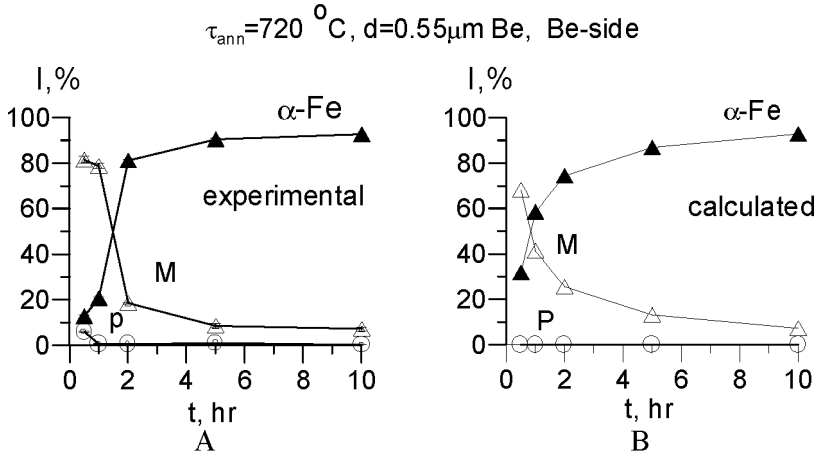

Fig.4 The experimental (A) and calculated (B) relative intensities of the partial CEMS-spectra in system Fe(11μm)-Be(0.55μm) versus the time of isothermal annealing at temperature of 720°C .

CONCLUSIONS

In this paper the results of investigation of the thermal-induced phase transformations in α-iron with the beryllium coating produced by magnetron sputtering after isothermal and isochronal were carried out are presented. The results are as follows:

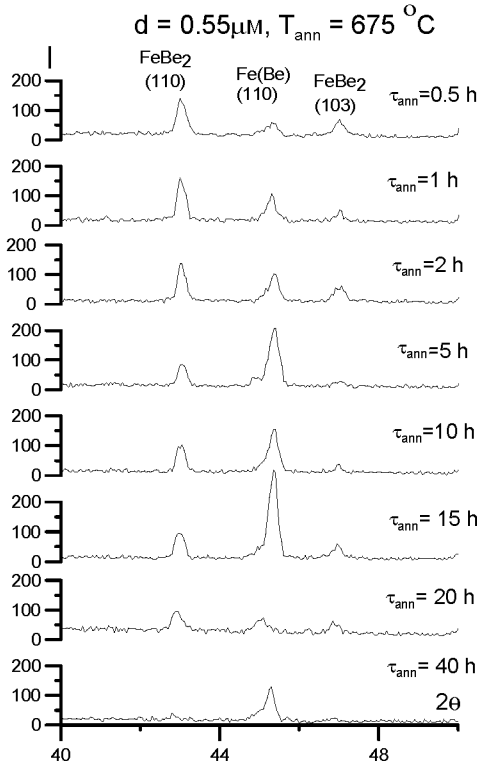

Fig.5 The X-ray diffraction patterns in binary system Fe(11μm)-Be(0.55μm) versus the time of isothermal annealing at temperature 675°C .

1) A succession of phase transformations in subsurface layers and in bulk begins from high order beryllides formation, proceeds by decreasing their order and completes by the solid solution Fe(Be) formation. As the thickness of coating grows, the main features of phase transformation processes ("competition" and disintegration of the beryllium-contained phases), take place at the higher annealing temperatures. The temperature increasing of isothermal annealing leads to shifting succession of phase transformations to law temperature region.
2) The phase formation rates essentially exceeds of over diffusion rates.
3) The physical model of diffusion and phase formation in the two-layer binary Fe-Be system has been proposed. The model takes into account the mutual diffusion mechanism, peculiarities of phase diagram Fe-Be.
4) The computer code that implements the proposed model has been developed. The code describes the phase formation kinetics in any region of the two-layer binary system at any regime of annealing .
5) Good agreement between the calculations and the Mössbauer investigations of both the interior and the subsurface layers of the two-layer binary system Fe-Be has been obtained.
6) The frequency factors and the energies of activation of iron in beryllium and beryllium in iron are in good agreement with appropriate literature data. The diffusion coefficients don't depend on a formed phase state in specimen depth.

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