Modelling of Mossbauer spectra of layered metal systems obtained by ion-plasma sputtering

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Abstract. The layered Zr(2 μm)-Fe(10 μm), Sn(4 μm)-Fe(10 μm) and Sn(4 μm)-Zr(2 μm)-Fe(10 μm) systems subjected to isochronous annealings have been studied. The sequence of phase transformations is determined. In the three-layer system, the formation of intermetallic compounds with a high content of Zirconium and a solid solution α-Fe(Sn, Zr) is established. The spectra of 57Fe nuclei in Iron-based alloys of various concentrations were calculated. A comparison with the experimental spectra of the layered systems obtained by ion-plasma deposition of Tin and Zirconium on an armco iron substrate and subjected to thermal annealing at 750 °C showed a good correlation.

1. Introduction

Equilibrium diagrams [1] of the binary systems Fe-Sn and Fe-Zr are characterized by the existence of regions of the solid solution α-Fe(Sn) and α-Fe(Zr) with various intermetallic compounds with increasing of temperature. In contrast to traditional methods, the ion-plasma deposition allows the formation of layers with high adhesion and the necessary chemical composition. High corrosion resistance in combination with mechanical strength, high melting point and low effective cross section for the absorption of thermal neutrons have recently made extensive use of Zircaloy (alloy of Zirconium with Iron and Tin) in reactor engineering. The study of thermally induced phase formation in two-layer systems Sn-Fe and Zr-Fe, as well as in three-layer systems Sn-Zr-Fe is relevant. Simulation of the phase state in layered systems will make it possible to predict changes during thermal treatment.

In this paper, the results of modeling the spectra of the phases formed in Zr-Fe, Zr-Sn-Fe and Sn-Fe layered systems, obtained by applying a Zirconium and Tin coatings on thin Iron foils by ion plasma deposition and subjected to thermal annealing are presented.

2. Experimental

The substrates for the studies were prepared from a bar of armco iron (99.8% Fe) by rolling on a roll to a thickness of \( d_{Fe} \approx 10\pm1 \) μm and subsequent homogenizing annealing at a temperature of 850 °C for 2 hours. The deposition of Zirconium and Tin on Iron foil substrates was carried out by method of ion-plasma sputtering. Thermal annealing of two-layer Zr(2 μm)-Fe(10 μm), Sn(4 μm)-Fe(10 μm) and three-layer Sn(4 μm)-Zr(2 μm)-Fe(10 μm) systems was carried out at 550–850 °C temperatures for 5 h in a vacuum surface with a residual pressure of \( 6\times10^{-6} \) mm Hg. Samples were studied by Mossbauer
spectroscopy (MS) method on $^{57}$Fe nuclei at room temperature. As a source of $\gamma$-quanta $^{57}$Co (Rh) with an activity of $\approx 5\mu$Ci was used.

The Iron-based layered systems are considered. Using the MSTools software package [2], the spectra of Iron nuclei were obtained for each position in the phases of binary systems, and taking into account the occupancy of the positions, the standard spectra of each phase were modeled. Next, using the lever rule, the spectra of alloys with different concentrations of the second component were calculated for the reference points of the state diagrams.

3. Results

The M"ossbauer spectrum of the layered Zr-Fe system in the initial state and up to a temperature of 600 °C is a Zeeman’s sextet. Thermal annealing at higher temperatures leads to the appearance of additional lines of the magnetically ordered phase with substantially smaller fields (figure 1a). Processing the spectra using the method of recovering the distribution functions of the magnetic field (figure 1b) showed the appearance of an intermetallic phase with parameters characteristic of the intermetallic compound Fe$_3$Zr [3].

![Figure 1. Experimental Mossbauer spectra of layered system Zr-Fe (a) and partial spectra of formed phases (b) after isochronous thermal annealing at various temperature.](image)

The Mössbauer spectra of the Sn-Fe layered system subjected to thermal annealing are presented in figure 2a. The positions of the lines for the spectra [4–7] of the various phases are shown. It can be seen that already at a temperature of 600 °C, a new phase is formed and an increase in temperature leads to significant phase transformations. As a result of processing, the spectra, the mechanism of phase transformations of intermetallic compounds (figure 2b) was revealed:

$$FeSn \rightarrow Fe_3Sn_2 \rightarrow Fe_3Sn \rightarrow Fe_5Sn_3.$$  

The Mössbauer spectrum of the Sn-Zr-Fe three-layer system, up to a temperature of 600 °C, remains unchanged (figure 3a). An increase in the annealing temperature leads to the appearance of magnetically ordered phases and phases of the paramagnetic type (the presence of lines in the center of
the spectra). The processing of the spectra showed the presence of intermetallic phases Fe$_3$Sn$_2$, Fe$_3$Sn, Fe$_5$Sn$_3$ and intermetallic compounds FeZr$_2$, FeZr$_3$ [8–9].

**Figure 2.** Experimental Mossbauer spectra of layered system Sn-Fe (a) and partial spectra of formed phases (b) after isochronous thermal annealing at various temperature.

**Figure 3.** Experimental Mossbauer spectra of layered system Sn-Zr-Fe (a) and partial spectra of formed phases (b) after isochronous thermal annealing at various temperature.
Using the phase diagrams of the binary systems Fe-Zr and Fe-Sn, according to the method [10], we simulated the Mössbauer spectra of two and three component alloys with different content of alloying components. Comparison with the experimental spectra of Zr-Fe, Sn-Fe and Sn-Zr-Fe layered systems subjected to isochronous 5 h annealing at 750 °C showed that the experimental spectra of the layered Zr-Fe, Sn-Fe, Sn-Zr-Fe systems correspond to model spectra of alloys Fe-10% Zr, Fe-15% Sn, Fe-10% (Zr-Sn).

**Figure 4.** Experimental Mossbauer spectra of Zr-Fe (a), Sn-Zr-Fe (e) and Sn-Zr-Fe (i) layered systems and calculated spectra of Fe-10%Zr (b), Fe-15%Zr (c), Fe-20%Zr (d), Fe-10%(Sn-Zr) (f), Fe-15%(Sn-Zr) (g), Fe-20%(Sn-Zr) (h), Fe-10%Sn (j), Fe-15%Sn (k), Fe-20%Sn (l) alloys.

4. Conclusions
As a result of the studies carried out, the sequence of the phase transformations in the layered Zr(2 μm)-Fe(10 μm), Sn(4 μm)-Fe(10 μm) and Sn(4 μm)-Zr(2 μm)-Fe(10 μm) systems that were subjected to isochronal annealings has been established. In the three-layer system, the formation of intermetallic compounds with a high content of Zirconium and a solid solution of Zirconium and Tin in Iron α-Fe(Sn, Zr) is established. The spectra of $^{57}$Fe nuclei in Iron-based alloys of various concentrations were modeled. A comparison with the experimental spectra of the layered systems obtained by ion-plasma deposition of Tin and Zirconium on an armco iron substrate and subjected to thermal annealing at 750 °C showed a good correlation.

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