Modelling of phase composition of Sn-Fe layered system obtained by ion-plasma sputtering

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Abstract. The Mössbauer spectra on Iron and Tin nuclei in different states in the phases of the binary system Fe-Sn are modeled. Reference spectra of intermetallic compounds and α-Fe (Sn) solid solution were obtained. The spectra of Iron and Tin alloys of various concentrations were calculated. A comparison with the experimental spectrum of the layered Sn(4 μm)-Fe(10 μm) system obtained by ion-plasma deposition of Tin on an armco iron substrate and subjected to thermal annealing at 700°C showed a good correlation.

1. Introduction

Ion technologies make it possible to obtain volume-inhomogeneous structures that are unattainable in the classical alloying of metals. Iron and Tin are components of zircalloys (zirconium-based alloys) used in reactor construction. To understand the processes in complex multicomponent systems, it is necessary to study the processes occurring in binary systems. According to the phase diagram, there are five intermetallic compounds in the alloys of the Fe-Sn binary system: Fe₂Sn, Fe₅Sn₃, Fe₃Sn₂, FeSn, Fe₅Sn₂ [1]. The β’-phase (Fe₅Sn), stable in the temperature range of 750–900°C, has a hexagonal structure of the type Mg₂Cd (a=5.448 Å, c=4.361 Å) [2]. The β'-phase (Fe₃Sn₂), stable in the temperature range of 600–815°C, has a monoclinic structure with a cell (a=13.53 Å, b=5.34 Å, c=9.20 Å and β=103°). The phase-phase (FeSn), which is stable below 745°C, has a hexagonal structure of the CoSn type (a=5.298 Å, c=4.446 Å). The FeSn₂ compound, which is stable below 496°C, has a tetragonal structure of the CuAl₂ type (a=6.533 Å, c=5.323 Å). On the phase diagram of the binary system Fe-Sn [1], there are also solid solutions α-Fe(Sn) and β-Sn(Fe). The phases have been investigated by the Mössbauer effect [3–9]. To analyze the experimental Mössbauer spectra, the information on the spectra of the different phases on ⁵⁷Fe and ¹¹⁹Sn nuclei at room temperature is necessary.

The sextets with hyperfine fields H₁=171±3 kGs, H₂=231±3 kGs and H₃=185±3 kGs and isomeric shifts δ₁=0.10±0.01 mm/s, δ₂=0.20±0.01 mm/s and δ₃=0.23±0.01 mm/s (rel. Cu) correspond to Iron atoms in positions 2(a), 2(d)-1 and 2(d)-2 for Fe₃Sn₃ [4]. The parameters of the intermetallic compound Fe₂Sn [5] were determined: δ=0.35±0.04 mm/s, δ=0.29±0.06 mm/s, H₆=254±5 kGs (on ⁵⁷Fe nuclei) and δ=1.45±0.20 mm/s, H₆=82.8±2.0 kGs (on ¹¹⁹Sn nuclei). Isomeric shifts are given relative to stainless steel 310 and SnO₂. For the ⁵⁷Fe nuclei in the compound Fe₃Sn₂ [6], the following parameters were determined: δ=0.57±0.01 mm/s (relative to α-Fe), δ=0.21±0.02 mm/s, H₆=196±3 kGs. It has been established [7] that the intermetallic compound Fe₃Sn₂ has the parameters: δ₁=1.92 mm/s, δ₂=1.62 mm/s, H₁=52 kGs; δ₁=1.79 mm/s, δ₂=1.04 mm/s, H₁=15.5 kGs (¹¹⁹Sn). The...
Iron atoms [8] occupy crystallographically equivalent 3(f) positions in the FeSn compound, while the Tin atoms occupy 1(a) and 2(d) positions in the ratio 1:2. Mössbauer parameters: for $^{119}$Sn nuclei – $\delta=1.97 \pm 0.05$ mm/s, $\Delta=1.72 \pm 0.10$ mm/s and $\delta=2.14 \pm 0.05$ mm/s, $\Delta=-2.82 \pm 0.10$ mm/s, $H_n=14.4 \pm 1.5$ kGs; for $^{57}$Fe nuclei – $\delta=0.68 \pm 0.02$ mm/s, $\Delta=-0.45 \pm 0.02$ mm/s, $H_i=120.9 \pm 1.5$ kGs, $H_f=113.6 \pm 1.5$ kGs, $H_y=101.4 \pm 1.5$ kGs. The parameters of the FeSn$_2$ intermetallic compound at $^{57}$Fe and $^{119}$Sn nuclei are as follows: $\delta=0.51 \pm 0.01$ mm/s, $H_n=113 \pm 1$ kGs; $\delta=2.21 \pm 0.02$ mm/s, $\Delta=0.86 \pm 0.02$ mm/s, $H_c=23.4 \pm 1.0$ kGs [9]. The Mössbauer spectrum of $\beta$-Sn is a singlet with $\delta=-0.69 \pm 0.01$ mm/s [10].

Thus, it is seen that nuclear gamma resonance spectroscopy is a very informative method. Using this method, it becomes possible to obtain simultaneously a wide range of data on the state of the electron shell of an atom, on the symmetry of the location of the surrounding charges, on the parameters of effective internal fields and on the dynamics of atoms.

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

2. Experimental

The phase diagram of the Fe–Sn binary system was taken as the object of study. A set of methods for processing and analyzing Mössbauer data, realized as the MSTools software package [11], was used in our work. Currently, the MSTools complex consists of ten programs designed to process and analyze both experimental Mössbauer spectra and their parameters. To achieve the goal of the work, a model interpretation of the Mössbauer spectra was used using a priori information about the object of study (SPECTR).

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 $d_0=10 \pm 1$ µm and subsequent homogenizing annealing at a temperature of 850°C for 2 hours. The deposition of Tin on Iron foil substrates was carried out by method of ion-plasma sputtering. Thermal annealing of two-layer Sn(4 µm)-Fe(10 µm) systems was carried out at a temperature of 700°C for 5 h in a vacuum surface with a residual pressure of $6 \times 10^{-6}$ mm Hg. Samples were studied by Mossbauer spectroscopy (MS) method on $^{57}$Fe and $^{119}$Sn nuclei at room temperature. As a sources of $\gamma$-quanta, $^{57}$Co (Rh) and $^{110m}$Sn (BaSnO$_3$) with an activity of $\approx 10$ μCi and $\approx 2$ μCi was used.

3. Results

Thus, to solve the problem of modeling a binary system, it is necessary to create models of intermetallic compounds (Fe$_5$Sn, Fe$_7$Sn$_3$, Fe$_5$Sn$_2$, FeSn, FeSn$_2$).

Using the SPECTR program, the partial spectra of Iron nuclei in various positions of the crystal lattice of various phases of the Fe-Sn binary system are created. Since Iron and Tin atoms occupy several positions in some intermetallic compounds, which correspond to 3 sextets, 2 sextets, sextet and doublet, the PHASAN program [11] was used to obtain the final version of the spectra of these phases. PHASAN allows for quantitative phase analysis using the spectra of reference samples. Based on the reference spectra of the $^{57}$Fe and $^{119}$Sn nuclei (in different positions of the phases), the spectra of the phases were simulated (figure 1). The positions of the lines of the partial spectra are shown in the figure.

Based on the equilibrium diagram of the Fe-Sn binary system, the ratio of phases at 700°C in systems with different (10–35%) Tin atoms contents were determined using the “lever rule”. Further, PHASAN program was applied to calculate the spectra of Iron alloys with various concentration of Tin (figure 2).

For comparison, the experimental spectra of the Sn(4 µm)-Fe(10 µm) layered system after thermal annealing at 700°C are given. It can be seen that the obtained model spectra of alloys correlate well with the results of experimental studies.
Figure 1. Simulated spectra of $^{57}$Fe (a) and $^{119}$Sn (b) nuclei in intermetallic compounds of Fe-Sn binary system.

Figure 2. Calculated spectra of $^{57}$Fe (a) and $^{119}$Sn (b) nuclei of Fe-Sn alloys and the experimental spectrum of Sn-Fe layered system.

4. Conclusions
As a result of the studies carried out, the spectra of $^{57}$Fe and $^{119}$Sn nuclei in Iron and Tin-based alloys of various concentrations were modeled. A comparison with the experimental spectrum of the layered
Sn(4 μm)-Fe(10 μm) system obtained by ion-plasma deposition of Tin on an armco iron substrate and subjected to thermal annealing at 700°C showed a good correlation.

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