STRUCTURAL CHANGES IN NICKEL ALLOYS SUBJECTED TO GAMMA IRRADIATION IN A GASEOUS MEDIUM

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The structuring action of γ rays from 60Co and 57Co sources on dilute nickel alloys containing elements with a high affinity for oxygen was studied. It was shown that after γ irradiation in the alloys at room temperature there is a prolonged redistribution of the elements which is accompanied by the precipitation of dispersed oxides.

In recent years an increasing number of researchers have turned to the study of structure formation processes in open systems [1]. Irradiated materials are open systems since they can exchange energy or matter with the surrounding medium.

In [2] it was shown that irradiation of iron and copper alloys by electrons and γ rays in different media allows one to change in their content of nonmetallic elements (C, S, N, H, O) over quite broad limits. The content of these elements can be both increased and decreased as a function of the irradiation conditions. The change of the element content may be accompanied by phase transformations.

Previously [3] it was found that the process of matter exchange with the surrounding gaseous medium occurs not only during γ irradiation but continues for some time after it has ceased.

In the present work, out study of the evolution of the alloy–gas system during its irradiation by γ rays of dose (0-5)·10¹⁷ Rad was carried out using as an example the dilute nickel alloy Ni plus 1 at. % ⁵⁷Fe; 1.3 at. % W; 1.15 at. % Al; 0.1 at. % Gd. It was found that there was a specific change in the alloy during γ irradiation and also continuing for a time after it had stopped. The study of the quantitative and compositional changes in the gaseous medium carried out independently of the structural investigations of the alloys will be published elsewhere.

MATERIAL AND INVESTIGATION METHOD

The alloy of the above-mentioned composition was prepared from NO grade nickel, iron enriched to 90% in the isotope ⁵⁷Fe, "very pure" grade tungsten, 99.999% aluminum, and GdM-1 grade gadolinium. The alloy was melted in a SSHVL furnace with secondary vacuum remelting and homogenization at 1573 K for 50 h in vacuum followed by slicing into 3 mm thick slabs.

The samples for irradiation were prepared from foils 10-30 μm thick produced by cold rolling with intermediate annealing in vacuum at 1273 K for 1 h or without it. During these anneals in the alloys studied containing elements with a high oxygen affinity, a zone of internal oxidation up to ~10 μm with dispersed oxides was formed [17]. The alloys were irradiated with a ⁶⁰Co source (E ≈ 1.27 MeV) at a dose rate of 156 R/sec and a ⁵⁷Co source (E ≈ 0.1 MeV) with an activity of 30 mC in air. We carried out a lengthy post-radiation soak of the alloy in air at room temperature.

The kinetics of the change in the alloy structure during its post-radiation soak were studied by Mössbauer spectroscopy of ⁵⁷Fe impurity atoms. The Mössbauer spectra were obtained using a YaGRS-4M spectrometer in a transmission geometry in the constant velocity regime with ⁵⁷Co in chromium as the source. The studies were done in air at room temperature. The spectra were analyzed using a program [4] for an IBM PC-286.

EXPERIMENTAL

During the investigations of structural changes in the γ-irradiated alloy, it was seen that depending on the prehistory of the treatment the initial alloy samples differed somewhat in composition and state. This was seen in the characteristics of the Mössbauer spectra and the effected structural changes during and after γ irradiation.

Figure 1 shows the Mössbauer spectrum of the alloy obtained by rolling with intermediate vacuum annealing at 1273 K for 1 h in the initial state 1 (IC1). The spectrum is the sum of several spectra obtained over 5 days. Besides the sextet \( H_1 \) from the main phase, it contained sextets with large fields \( H_2 , H_3 , \) and \( H_4 \). The expansion of this spectrum into four sextets with the usual ratios of the positions of the six components and relative intensities 3:3:1:1:3:3 gave the characteristics given in Table 1.

It can be seen that the line width in sextet \( H_1 \) is substantially larger than the natural linewidth of 0.194 mm/sec, and thus description of it by a single sextet is insufficient. This spectrum should be regarded as a superposition of yet at least two or more spectra, which are formed by different local configurations of the surroundings for the \( ^{57}\text{Fe} \) atoms. The same can be said about sextet \( H_4 \).

Immediately after irradiating with \( \gamma \) rays, the sextets \( H_3 - H_4 \) with large fields vanish, while the linewidth of sextet \( H_1 \) increases (Fig. 2).

An increased width is also observed during a prolonged postradiation soak of the alloy in air at room temperature. Table 2 shows the characteristics of the overall spectra of the samples if each of them represents a single sextet.

With a long soak in air at room temperature, a redistribution of the components of the alloy occurs in the sample leading not only to broadening of the sextet \( H_1 \) but also the appearance of sextets with large fields \( H_{2-3} \) and \( H_{3-2} \) (Fig. 3). This is most clearly seen in spectra obtained with an extended velocity scale. The expansion of the spectrum into three sextets with an intensity ratio 3:3:1:1:3:3 gave the values shown in Table 3.

It can be seen that the first and third sextets are broadened and it follows to represent them as a superposition of a large set of sextets which correspond to a large number of inequivalent positions of the \( ^{57}\text{Fe} \) atoms in the phase or different in its surroundings in the solid solution, which shows up during prolonged postradiation soak of the sample in air. A further soak (2 months after irradiation) can lead to shifting of the outer lines of the \( H_{2-3} \) sextet opposite to each other (average hyperfine magnetic field \( H = 453 \) kOe) and a decrease of their width.

It should be noted that the appearance of a series of sextets with large fields in the spectrum of an alloy is also seen with another form of preliminary treatment and irradiation. Figure 4 shows the resonant absorption spectrum of an alloy sample produced by cold rolling (\( \varepsilon \approx 99.8\% \)) without intermediate annealing. Besides the broadened sextet with \( H_1 = 256.5 \) kOe, \( \delta = -0.283 \) mm/sec, \( \Delta = 0.012 \) mm/sec, and \( \Gamma_1 = 0.42 \) mm/sec, the spectrum also contained a small paramagnetic component. If we represent it by a single line then it has \( \delta = -0.11 \) mm/sec and \( \Gamma = 0.19 \) mm/sec, and has area ~3%.

The Mössbauer spectrum of this sample was also taken after low energy \( \gamma \) irradiation of it in air and then in argon at room temperature and a following soak in air for 12 days. The spectrum was taken in the course of one day and is shown in Fig. 5. A new phase with strongly broadened sextet lines appeared.

DISCUSSION OF RESULTS

For \( ^{57}\text{Fe} \) atoms in pure nickel at 4 K [5], the following parameters have been reported for the Mössbauer spectrum: \( H_{\text{eff}} = 283 \pm 0.03 \) kOe and \( \delta = 0.12 \) mm/sec. At room temperature for \( ^{57}\text{Fe} \) atoms implanted in nickel (fcc structure), a similar magnetic field was obtained [6] and also \( H = 320 \) kOe for \( ^{57}\text{Fe} \) nuclei introduced by alloying or diffusion. The dependence of the average hyperfine magnetic field on the \( ^{57}\text{Fe} \) nuclei on the nickel concentration in the alloys of the Fe–Ni system can be found in [7].

On the basis of all of this data and also the assertions of [5, 8, 9] that in dilute solid solutions the presence of nickel atoms in the nearest surroundings of \( ^{57}\text{Fe} \) atoms only weakly changes \( H \), we can suppose that in the alloys under study none or very few of the \( ^{57}\text{Fe} \) atoms are surrounded exclusively by nickel atoms. Only a small amount of \( ^{57}\text{Fe} \) is present in the neighborhood of Fe atoms (5.6%); they have a field \( H = 333 \) kOe). The remaining \( ^{57}\text{Fe} \) atoms in the solid solution evidently must have nearest neighbors of impurities besides Ni.