Effect of heterogeneous distributed intermetallic precipitates on accumulation of vacancy-like defects in irradiated Fe-Ni-based alloys studied by positron annihilation

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Abstract. The effect of heterogeneous distributed intermetallic particles on interaction of point defects with dislocations in cold-worked Fe-Ni-based alloys during electron irradiation was studied by positron annihilation. It was shown that vacancy accumulation occurs in cold-worked alloys during irradiation in spite of high dislocation density. It is caused by the presence of areas with lower dislocation density in alloys. At room temperature, the vacancies are not trapped by dislocations totally because a low their mobility. At elevated temperatures, vacancies may leave dislocations and form VCs. In cold-worked Fe-Ni-Ti alloy the accumulation of defects during irradiation is essentially lowered in comparison with cold worked Fe-Ni alloy. Ni$_3$Ti precipitates, which are present on dislocations, reduce the efficiency of interaction of dislocations with interstitial atoms and, thereby, enhance the mutual recombination of point defects.

4. Introduction

Irradiation-induced swelling is a main limiting factor for using of austenitic stainless steels as structural materials of critical core components of reactors. The swelling is due to a vacancy accumulation occurred under irradiation that is caused by bias factors of various sinks [1]. The preliminary plastic deformation reduces effectively the vacancy accumulation because dislocations, which are formed during deformation, are sinks for vacancies [1-3]. However, a locally inhomogeneous microstructure is formed in austenitic alloys and steels during deformation. Processes of the radiation damage in materials having an inhomogeneous distribution of dislocations are not quite clear. Moreover, an initial dislocation structure substantially changes during irradiation that is caused by dislocations motion (climb and glide). As a result, the effect of deformation drops after irradiation to high doses (∼50 dpa).

The dislocation structure may be stabilized by ultrafine intermetallic precipitates which form in aging alloys. These precipitates strongly pin dislocations and thereby block a dislocation motion [4]. Moreover, if they are located on dislocation lines, the precipitates may affect the interaction of point defects with dislocations [1]. However, experimental data about investigations of the effect of intermetallic precipitates on interaction of dislocations with point defects is absent today.

In this paper, we present the results of investigation of the vacancy defects evolution during irradiation of Fe-Ni-based alloys in various initial states (solution annealed, cold-worked) by the positron annihilation spectroscopy. These alloys were used as model materials for fast breeder reactor
stainless steels. Defects were induced by electron irradiation, which generated homogeneously
distributed Frenkel pairs. The freely migrating point defects (interstitial atoms and vacancies) are only
formed at electron irradiation unlike it at the neutron irradiation. The study of interaction of defects
with sinks, alloying additions and over features is able in alloys in case of electron irradiation.
Experimental results of our research were also published earlier [5].

5. Materials and methods
Samples of Fe–36.5 Ni (Fe-Ni) and Fe-36.5 Ni-2.5 Ti (Fe-Ni-Ti) fcc alloys (wt %) were used. After
rolling, cutting and electrical polishing, the samples were solution annealed under a 10^-6 Pa vacuum at
1373 K for 1 h and then were cooled quickly (the SA state). Some alloy samples were deformed by
rolling to 40% at room temperature. After cold-working, alloys were thermally treated at 650 K for 1 h
in order to anneal the vacancy defects formed during deformation (the CW state) [4]. The
microstructure of the solution annealed and deformed samples were certified using by transmission
electron microscopy (TEM). The samples of the SA alloys had grains ~30 µm in size and the
dislocation density of about 10^{11} m^{-2}. In samples of CW alloys a banded subgrain structure was
formed during deformation. The subgrains, which were extended lengthwise, were 50 to 400 nm wide.
Dislocations in subgrains formed cellular-network configurations. The mean dislocation density inside
subgrains was about 9x10^{14} m^{-2} [4].

The samples were irradiated at 300 and 573 K with 5 MeV electrons in a linear accelerator. The
maximum electron fluence was 5x10^{22} m^{-2}, which corresponded to the damaging dose of 5x10^{-4} dpa
as calculated by the modified Kinchin-Pease model [6].

The angular correlation of annihilation radiation (ACAR), which is a one of positron annihilation
methods, is used to investigate of defect structure. ACAR measurements were performed at room
temperature. The ACAR method was realized in a one-dimensional ACAR spectrometer providing a
resolution of 1 mrad x 160 mrad. A ^{68}Ge positron source of activity of 400 MBq was used. At least
8x10^5 coincidence counts were collected in each ACAR spectrum; the peak-to-background ratio was
~10^3. The ACAR spectra of the alloys were approximated by an inverted parabola and a Gaussian. The
approximation quality criterion approached unity [7]. Changes in the shape of the ACAR spectra were
characterized by the standard S-parameter which were defined as the ratios of low-regions (p < 3.5x10^{-3} m_0c)
to the total region [8].

6. Results and discussion

6.1. Irradiation at room temperature
The Figure 1 presents the dependence of the S-parameter on the electron fluence for Fe-Ni and Fe-Ni-
Ti alloys in SA and CW states irradiated at 300 K. The detailed description of results was reported in
our recent work [5]. We have been estimated from our results a concentration of vacancies presented
in alloys after irradiation to various fluencies. It should be noted that high dislocation density (about
9x10^{14} m^{-2}) is preserved inside subgrains in CW alloys after deformation and annealing [4]. Therefore,
we use a Simple Trapping Model (STM) in our estimations. According to three positron state model,
the S-parameter is related to the concentration of vacancies presented in samples C_0 as [9]:

\[ C_0 = \frac{\lambda_f (S - S_f) + \mu_d \rho_d (S - S_d)}{\mu_v (S_j - S)} \]

where \( \lambda_f \) is the positron annihilation rate in the bulk (free) state; \( \mu_d \) and \( \mu_v \) are the specific positron
trapping rates for dislocations and vacancies respectively; \( S_f, S_d \) and \( S_j \) are the S-parameters
characteristic of the positron annihilation from the bulk (free), dislocations and vacancy defect-trapped
states respectively. The next values of annihilation characteristics, which we obtained in [4,8,9], was
used for calculation: \( \lambda_f = 9x10^8 \text{ s}^{-1}, \mu_d = 1.2x10^{-4} \text{ m}^2/\text{s}, \mu_v = 2.2x10^{15} \text{ s}^{-1}, S_f = 0.526, S_d = 0.574, S_j = 0.605. \) It should be noted that the initial value of S-parameter for Fe-Ni-Ti(CW) alloy is much smaller
than $S_n$. As we shown earlier [4], in the Fe-Ni-Ti alloy, the fine intermetallic precipitates of Ni$_3$Ti-type located at dislocations. These precipitates reduce the trapping efficiency of positrons by dislocations. The estimates according trapping model give specific positron trapping rates for dislocation in Fe-Ni-Ti(CW) alloy $\mu_{d}^{Ti} = (2.1\pm0.3) \times 10^{-5} \text{ m}^2/\text{s}$.

As we shown earlier [10], vacancies are mobile in this alloy at room temperature and they form small (3-5 vacancies) three-dimensional clusters (VC). In this case, the $C_0$, which we obtain from Equation 1, correspond to concentration of vacancies bounded in VCs. The results of calculation are shown in Figure 2.

As it can be seen from figure, the vacancy accumulation occur in Fe-Ni(SA) during irradiation. In Fe-Ni-Ti(SA), alloy the vacancy accumulation is enhanced in comparison with that in Fe-Ni(SA) alloy. Ti is an over-sized impurity to the Fe-Ni model alloys. Ti atoms interacts with migrating vacancies that result in enhancement of vacancy accumulation [4,5,10].

In Fe-Ni(CW) alloy, the vacancy accumulation is reduced in comparison with that in Fe-Ni(SA) alloy at higher fluencies ($\Phi \geq 2 \times 10^{22} \text{ m}^{-2}$). However, the difference between vacancy concentration in SA and CW alloys is very low ($\approx 1.5$ times) in spite of a high density of sinks presented in cold-worked alloy. As it mentioned above, the dislocations in subgrains formed cellular-network configurations. The dislocation density inside cells is one order of magnitude smaller (about $10^{14}$ m$^{-2}$) than mean density of dislocations. The diffusion length of vacancies is very low at this temperature (a few nanometers) and a deal of vacancies doesn’t reach the dislocations [11]. As a result, these vacancies are not absorbed by sinks and form VCs. It should be noted that in fcc metals interstitial atoms have high mobility and they are absorbed totally by dislocations [8].

In Fe-Ni-Ti(CW) alloy, the vacancy accumulation is surprisingly much smaller than that in over alloys. The investigations of annealing of irradiated alloys had shown [4,5] that the Ti-vacancy interaction takes place also in this alloy. Thus, a vacancy accumulation in Fe-Ni-Ti(CW) alloy must be larger than that, at least, in Fe-Ni(CW) alloy. The most probable reason for reduction of vacancy accumulation is enhanced mutual recombination of point defects. It seems that precipitates, which are located at dislocations, reduce the efficiency of interaction of dislocations with interstitial atoms [1]. As a result, some number of interstitials is not absorbed by dislocations and recombines with vacancies.

6.2. Irradiation at 573 K

The Figure 3 presents the dependence of the S-parameter on the electron fluence for Fe-Ni and Fe-Ni-Ti alloys in SA and CW states irradiated at 573 K. The detailed description of results was reported in our recent work [5]. The dependence of concentration of vacancies presented in alloys after irradiation on the electron fluence is shown in Figure 4.

![Figure 1](image1.png)  
**Figure 1.** S-parameter variation versus irradiation fluence at 300 K for investigated alloys.

![Figure 2](image2.png)  
**Figure 2.** The vacancy concentration variation versus irradiation fluence at 300 K for investigated alloys.
As it can be seen from figure, in Fe-Ni(CW) and Fe-Ni-Ti(CW) alloys the vacancy accumulation occurs only at higher fluencies ($\Phi \geq 2 \times 10^{22}$ m$^{-2}$). However, the vacancy concentration in these alloys at fluence $\Phi = 5 \times 10^{22}$ m$^{-2}$ is more than that in Fe-Ni(SA) alloy. The vacancy mobility is high at this temperature and vacancies easily reach the dislocations and are trapped by them. However, as it shown in [11], trapped vacancies may leave dislocation core at elevated temperatures. At this, detrapped vacancies form VCs at higher fluencies, when their concentration is sufficiently high.

It should be noted that vacancy accumulation is reduced in Fe-Ni-Ti(CW) alloy in comparison with that in Fe-Ni(CW) alloy. Thus, the precipitates, which are located at dislocations, enhance the mutual recombination of point defects, as it takes place during irradiation at room temperature.

**Figure 3.** S-parameter variation versus irradiation fluence at 573 K for investigated alloys.

**Figure 4.** The vacancy concentration variation versus irradiation fluence at 573 K for investigated alloys.

### 7. Conclusions

In this paper we present results of ACAR characterization of electron irradiated Fe-Ni and Fe-Ni-Ti alloys with varied initial structure. It was found that vacancy accumulation occurs in cold-worked Fe-Ni alloy during irradiation in spite of high dislocation density. It is caused by the presence of areas with lower dislocation density in alloys. At room temperature, the vacancies are not trapped by dislocations totally because a low their mobility. At elevated temperatures, vacancies may leave dislocations and form VCs. In cold-worked Fe-Ni-Ti alloy the accumulation of defects during irradiation is essentially lowered in comparison with cold worked Fe-Ni alloy, Ni$_3$Ti precipitates, which are located at dislocations, reduce the efficiency of interaction of dislocations with interstitial atoms and, thereby, enhance the mutual recombination of point defects.

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