Helium accumulation in \( \text{Cr}_{18}\text{Ni}_{10}\text{Ti} \) austenitic steel under different saturation methods

S O Akayev\(^*\)\(^1\), G Z Ganeev\(^1\), A S Dikov\(^1\), S B Kisilitsin\(^1\), I V Khromushin\(^1\), Ya Partyka\(^2\)

\(^1\)Institute of Nuclear Physics, Ibragimov st.1, 050032 Almaty, Kazakhstan
\(^2\)Lublin University of Technology, NadbystrzyckaSt. 38 D, 20 – 618 Lublin, Poland

*E-mail: s.akayev@inp.kz

Abstract. Investigation results of changes in the surface structure and helium accumulation in steel \( \text{Cr}_{18}\text{Ni}_{10}\text{Ti} \) at various saturation methods are presented. It is shown that different methods of helium introduction can lead to both different and identical changes in the structure and to forms of helium accumulation.

1. Introduction

As is known, helium strongly affects the properties of structural materials for fission and fusion reactors [1-2]. This led to highlight the separation of the problem of helium in metals into a special subsection of the physics of radiation damage and radiation materials science, as well as to an intensive study of the behavior of helium in various metals and alloys. The level of material saturation with helium and the forms of its accumulation determine helium effect on the structure and physical and mechanical properties [3]. Helium effects on the structure and properties are varied: helium implantation on accelerators leads to blistering and flaking, the accumulation of transmutant helium in neutron-irradiated structural materials of fission and fusion reactors leads to their embrittlement and enhanced swelling.

The level of saturation with helium depends on the way of its introducing into material. According to the literature, the following methods are most often used to saturate materials with helium:

- Ion implantation on charged particle accelerators, leading to surface erosion, blistering and flaking [4-6].
- Accumulation of transmutant helium due to nuclear reactions under neutron irradiation, which results in embrittlement, increased swelling and creep [7-8].
- Exotic methods of defect-free helium injection into studied material, which also leads to a change in properties [9].

Experimental studies the level and forms of helium accumulation mainly carried out by the methods of thermal desorption spectroscopy (TDS), using the results of computer modeling for the analysis of experiments: binding energies of helium atoms with crystal lattice defects (vacancies, dislocations, etc.), the activation migration energies helium-vacancy complexes. In this work, we have undertaken studies of the helium accumulation by TDS and surface structure investigations of \( \text{Cr}_{18}\text{Ni}_{10}\text{Ti} \) austenitic steel after various methods of helium saturation. Namely: by implantation at accelerator, by means of transmutation under irradiation with fast neutrons and under neutron irradiation in helium atmosphere [10]. The aim of the work is to identify general trends and differences under different methods of saturation of steel with helium.
2. Material, samples preparation, irradiation procedure and investigation methods

How already mentioned above, steel Cr$_{18}$Ni$_{10}$Ti was chosen as material for investigation. This steel widely uses as structural material of nuclear facilities.

Studies of the effect of helium accumulation on the surface structure implemented for three methods of introducing helium into steel.

The first method was helium implantation on accelerators. Irradiation was carried out on the low-energy channel of the DC-60 accelerator with doubly charged helium ions with energy of 22.5 keV/charge, that is the total charge of $\alpha$-particles was 45 keV. Irradiation was performed at room temperature (300K). The fluence of $\alpha$-particles was $1 \times 10^{16}$, $5 \times 10^{16}$, $1 \times 10^{17}$, $1 \times 10^{18}$ cm$^{-2}$. The projective range of $\alpha$-particles with energy of 45 keV in steel is ~ 150 nm, the straggling is ~ 70 nm.

For the second method of introducing helium, an ampoule of Cr$_{18}$Ni$_{10}$Ti steel filled with helium was designed and manufactured. Irradiation of the capsule filled with helium was carried out in the vertical channel of the WWR-K research reactor (INP, Almaty). The total irradiation time was 3254 h 52 min. The neutron flux during irradiation was: $7.3 \times 10^{13}$ n/(cm$^2$·s) for neutrons with $E \leq 0.1$ MeV (thermal neutrons) and $6.8 \times 10^{12}$n/(cm$^2$·s) for neutrons with $E \geq 0.1$ MeV, that is for "fast" neutrons. During irradiation, the samples for research received fluence of $8.5 \times 10^{20}$ n/cm$^2$ for thermal and $8.9 \times 10^{19}$ n/cm$^2$ for fast neutrons. Irradiation temperature was 993K. Samples for research were cut from the walls of the ampoule and prepared for further study.

The third method is the accumulation of helium due to nuclear reactions during the interaction of neutrons with structural material in the core of the BN-350 reactor. As samples for the study, we used samples cut from the face of hexagonal shroud of the N-214 spent fuel assembly (SFA) at different elevations along the assembly height relative to the center of the reactor core. The irradiation dose of the two studied samples was ~ 15 dpa (specimen No. 1 in Figure 1,c), and ~ 1 dpa (specimen No. 2 in Figure 1, c). Irradiation temperature of two samples was 610K and 554K, respectively. SFA, from which samples were cut out were kept for definite times in a wet storage after irradiation. Samples for research were stored in a dry storage of radioactive materials ~ 10 years. During storage, the plates were in unstressed state.

Registration of helium in thermal desorption experiments was fulfilled by using an MX-7304 radio-frequency mass spectrometer. Thermal desorption curves were automatically recorded during heating in a vacuum of ~ $1.3 \times 10^{-6}$ Pa at a rate of 42 K/min in the range of 300 - 1373K. Before measurements, a calibration was performed for the helium content in the vacuum chamber. To study the desorption of helium by the method of thermal desorption spectroscopy, samples with dimensions of 3×2×1 mm were prepared.

The study of the steel surface structure was carried out by scanning electron microscopy (SEM) on Hitachi TM4000 microscope.

3. Results and discussion

Investigations accumulation of helium after various methods of its introduction was carried out by the method of thermal desorption spectroscopy.

The thermal desorption spectrum of helium implanted by irradiation of steel with $\alpha$-particles is shown in Figure 1 (a). The spectrum contains four peaks of helium yield, first at 520K, second – 600K, third – 660K, forth – 800K. The main peak yield is observed at 800K. At a fluence of $1 \times 10^{18}$ cm$^{-2}$, a hillocks appear on the steel surface (Figure 2, a), which are a manifestation of filled with helium subsurface bubbles. Some of the blisters have opened up to the surface. This means that the critical fluence for blisters formation has been reached. Blister cover is destroyed when their dimensions become 5 μm and more, blisters with smaller dimensions are not destroyed. Sequential annealing in the temperature range 400–800K leads to an increase in the size of the blisters due to the coalescence of small blisters and migrating helium atoms trapping. The low-temperature peaks of desorption in Figure 1, a indicate that, simultaneously with coalescence, the process of helium release from vacancy-helium clusters and its migration to the surface occurs. At annealing temperature of 800K, the number of blisters on the surface noticeably decreased, but their size increased due to the
coalescence with small blisters and helium atoms trapping. In most blisters the cover destroyed. At a temperature of 1000K, the cover destroyed in almost all blisters.

Figure 1. The spectrum of thermal desorption of helium from steel Cr18Ni10T after irradiation with helium ions at accelerator (a), neutron irradiation in helium atmosphere in research WWR-K reactor (b), irradiation in the BN-350 fast neutron reactor (c). 1 and 2 on figures a, b, c means numbers of investigated specimens: No 1 and 2

For steel irradiated with neutrons in a helium atmosphere at temperature of 993K, the maximum helium yield is achieved at a temperature of ~ 500K (peak II). Peaks I and III correspond to temperatures of 450K and 590K, respectively, see Figure 1b. Note that the yield of helium in TDS experiments in this case is much lower than in the previous case of irradiation at an accelerator. The distribution of implanted helium and point defects under high-temperature irradiation with neutrons in a helium atmosphere is fundamentally different from the previous case of irradiation with monoenergetic α-particles at low temperature ~ 300K. The spectrum of the WWR-K reactor (irradiation channel 10-2) is dominated by thermal neutrons with energy of ~ 15 keV [11], which do not create point defects. Helium atoms inject into steel due to interaction with neutrons is concentrated near the surface with maximum at a distance of ~ 20 nm from it. Due to the high (> 0.5Tmel) irradiation temperature, an almost uniform distribution of thermal vacancies is created in the bulk of the steel. During irradiation most of the highly mobile helium atoms desorbs from the surface, the surviving helium atoms accumulate in clusters, which consist of vacancy + helium atom and uniformly distributed over the steel bulk and trapping by dislocations. Such complexes have a high binding energy and helium "survives" in them up to temperature 600K. This can explain the third peak of the helium yield. It is noteworthy that the helium yield temperature (590K) and the calculated activation energy of the third peak see Table 1, are close to helium yield temperature (600K) and the activation energy for low-temperature helium implantation on an accelerator. The activation energies in Table 1 were calculated using the Redhead formula [9, 12]:

$$E = RT\left(\ln \frac{T_p v_1}{\beta} + \ln \frac{1}{\ln \left(\frac{T_p v_1}{\beta}\right)}\right),$$

Where R is the universal gas constant, $T_p$ is the peak temperature, β is the heating rate. The frequency $v_1$ is taken equal to $10^{13}$ s⁻¹ [13].

Comparable values activation energies of helium yield for such different methods of helium introducing indicate the same mechanism of helium desorption, i.e. release of helium bound by dislocations or He atoms yield from HeV complexes (helium atom in vacant site of cristal lattice). Peaks I and II are apparently associated with the release of helium accumulated at the triple junctions of grain boundaries. The surface of the sample has no traces of blistering (Figure 2, b), however, transmission electron microscopy (TEM) made it possible to reveal single voids (gas bubles) at the grain boundaries in the bulk of steel, see Figure 3, a.
Table 1. Temperatures and activation energies for helium desorption

| Peak, No | Irradiation with α-particles | Neutron irradiation in helium atmosphere | Ducts of spent fuel assemblies |
|---------|-----------------------------|----------------------------------------|-------------------------------|
|         | I   | II  | III | IV | I   | II  | III | I   | II  | III |
| Temperature, K | 520 | 600 | 660 | 800 | 450 | 500 | 590 | 600 | 650 | 680 |
| Activation energy, eV | 1.47 | 1.71 | 1.88 | 2.3 | 1.28 | 1.45 | 1.78 | 1.72 | 1.87 | 1.94 |

Figure 2. Surface of samples: after irradiation with helium ions (a), neutron irradiation in a helium atmosphere (b), fast neutron irradiation in the BN-350 reactor (c).

The third considered in this work method is the accumulation of helium in structural materials during the operation in reactor. For example, at operation of fast neutron reactors are accumulation of transmutante helium generates in FA materials. According to [7], the production of helium depends on neutron irradiation dose and is ~ 1 ppm/dpa. As mentioned above, helium affects material structure and properties: it stabilizes vacancy complexes, promotes swelling, and initiates helium embrittlement. Obtained by TDS method the results on helium accumulation for samples of steel Cr_{18}Ni_{10}Ti irradiated up to various damage doses are shown in Figure 1, c. Figure 1 shows that the yield of helium from steel irradiated to dose of 15 dpa is much higher than that of steel irradiated to dose of 1 dpa. This confirms the validity of the relationship between helium generation and the dose of neutron irradiation.

Figure 3 CTEM images of gas bubbles in Cr_{18}Ni_{10}Ti steel after neutron irradiation in helium atmosphere (a), microstructure of Cr_{18}Ni_{10}Ti steel samples irradiated with fast neutrons in BN-350 reactor up to damage dose 15 dpa (b) and 1 dpa (c).

Irradiation with fast neutrons leads to almost uniform formation of point defects and generation of transmutante helium in the bulk of the material. The temperature dependence of the helium yield also shows peaks at 600K, both in steel irradiated up to 15 dpa and in steel irradiated up to 1 dpa. That is, a
peak in the yield of helium at 600K is present both for different methods and for different temperatures of helium injection into steel. Based on the works of the authors [14, 15], we can say that the peak at 600K is associated with the release of trapped helium at edge dislocations (activation energy is of order 1.8-2.3 eV), as well as from complexes of vacancy with a helium atom. The binding energies of helium atoms in such complexes are close to ~ 2 eV.

The surface of the samples irradiated with fast neutrons has no blistering traces (Figure 2, c), like to previous case neutron irradiation in helium atmosphere. Figure 3 (b) shows TEM image of a sample after irradiation to damage dose 15 dpa; one can see a large number of radiation-induced voids. Voids is absent in sample irradiated to dose of 1 dpa (Figure 3,c).

4. Conclusion
Helium accumulation in steel Cr$_{18}$Ni$_{10}$Ti under various saturation methods, namely: at α-particles implantation on accelerator, neutron irradiation in helium environment, generation of transmutant helium under irradiation with fast neutrons was studied by TDS, SEM, and TEM methods.

Formation of blisters occurs only upon implantation of helium ions at accelerator. Critical fluence of blister formation upon irradiation at room temperature is $5 \times 10^{17}-10^{18}$ cm$^{-2}$.

SEM and TEM studies have shown that irradiation with neutrons in helium atmosphere and fast neutrons leads to the accumulation of helium in the bulk of the steel, but do not lead to the formation of blisters on the surface.

As shown by TDS spectroscopy studies, despite the cardinal differences in the methods of helium incorporation, spectra’s of thermal desorption for all methods contains peak at temperature of ~ 600 K with an activation energy of helium yield 1.7-1.8 eV. This indicates that part of the embedded helium is accumulates in traps of similar nature despite of saturation methods. These can be helium trapped at dislocations or helium in vacant site of the crystal lattice.

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