The effect of neutron irradiation in a helium atmosphere on the mechanical properties of 18Cr10NiTi austenitic steel

S O Akaev¹,², A S Dikov¹,³, L A Dikova¹, S B Kislitsin¹,³, V V Firsova¹, A S Larionov¹, I I Chernov³ and M S Staltsov³

¹ Institute of Nuclear Physics of the Republic of Kazakhstan, Ibragimov st. 1, 050032 Almaty, The Republic of Kazakhstan
² Satbayev University, Satpaev st. 22a, 050013 Almaty, The Republic of Kazakhstan
³ National Research Nuclear University MEPhI (Moscow Engineering Physics Institute), Kashirskoe highway 31, 115409 Moscow, Russia

E-mail: lexa_edji@mail.ru, i_chernov@mail.ru

Abstract. The microstructure and mechanical properties of 18Cr10NiTi austenitic steel were studied after high-temperature neutron irradiation of samples in a helium atmosphere. It was revealed during irradiation, helium penetrates into steel samples forming bubbles distributed on dislocations and along grain boundaries. Character of the temperature dependence of the mechanical characteristics of irradiated steel indicates a high-temperature radiation embrittlement (HTRE) of steel. The character of the mechanical properties temperature dependence change of irradiated steel points to its high-temperature radiation embrittlement.

1. Introduction

It is known that the presence of helium in structural materials is one of the factors limiting their working capacity in nuclear reactor core [1–3]. One of such manifestations may be the formation of bubbles chains along grain boundaries. This leads to weakening of intergranular bonding and grain boundary cracking known as the phenomenon of high-temperature radiation embrittlement (HTRE) [1, 4, 5]. Along with this, there is a point of view according to which the effect of helium on the mechanism of HTRE is secondary, and it connects HTRE with the segregation of impurity atoms, hardening of the material inside the grains themselves as a result of gas bubbles formation, presence of vacancy clusters, particles of the secondary phases, as well as, possibly, by braking of polygonization and recrystallization processes. As a result, the imbalance in the strength of the grain body and its boundaries increases, leading to grain-boundary destruction of a irradiated material [6–8]. These authors justify the denial of predominant role of helium in a metals high-temperature ductility decrease by the fact that HTRE is most pronounced in materials that are able to loss of high-temperature ductility as a result of various treatments even without irradiation [6, 8]. For example [6], in the experiments of helium saturation by a defect-free method using the “tritium trick” method, the role of helium was not established, and the authors considered that HTRE is associated with an impurities state change in the metal and formation of precipitates under irradiation. However, the fact of the effect of helium on high-temperature embrittlement of materials was revealed unambiguously using a similar technique for introducing of helium into 304L and 309S steels, vanadium based alloy, niobium and its alloys and titanium alloys [5]. Despite of different views on the HTRE mechanisms, most researchers
attribute it to the influence of helium atoms formed in the reactor during irradiation \([1–3, 5, 9–12]\). It was found in these works the effect of helium on HTRE is significant, and helium is determining in some cases, and the main mechanism of this effect is the accumulation of helium at elevated temperatures at the boundaries in the form of bubbles chain.

Thus, it is not always easy to differentiate the effect of helium and other processes on the mechanism of HTRE. This is due to the fact that the helium accumulation rate correlates with the rate of radiation defects generation in steels, which, in turn, affect radiation-stimulated processes, including diffusion and segregation phenomena \([13, 14]\). It is known only for certain that the HTRE phenomenon manifests itself only in polycrystallines at temperatures above 0.45 of melting temperature \([2, 5]\).

Ion implantation methods and a tritium trick are widely used in addition to high-dose neutron irradiation to study the effect of helium on the properties of various materials \([2, 5, 15, 16]\). However, these methods do not allow to unambiguously isolate the role of helium in HTRE also. In addition, when using various methods to simulation of the HTRE process, there are a number of problems: high radioactivity of the irradiated material (neutron irradiation); small depth of helium penetration (ion-implantation of helium); generation of a large number of radiation defects (simulating irradiation by heavy ions with the simultaneous introduction of helium ions); hydrogenation, which also leads to embrittlement of the material (tritium trick).

Therefore, it is of interest to consider a helium accumulation in structural materials under high-temperature neutron irradiation during high-temperature neutron irradiation in a helium atmosphere up to damaging dose of \(~1\) dpa, i.e. at relatively low concentrations of introduced radiation defects.

2. Experimental procedure

The material for research was the austenitic steel 18Cr10NiTi in the state of 10% cold-worked (CW). This type of steel is widely used in nuclear reactors as a structural material for nuclear reactor internals. Investigation of helium penetration into steel during high-temperature neutron irradiation was carried out using gas-filled samples (ampoules). The ampoule wall thickness was 2 mm; the helium pressure during irradiation did not exceed atmospheric pressure. The irradiation temperature was 1033 K. Warming-up of ampoules was carried by neutron irradiation only. There was no contact of the ampoules with coolant during irradiation. Neutron irradiation was performed in the WWR-K reactor up to fluence of \(9 \times 10^{23} \text{n/m}^2\) at neutron flux (\(E > 0.1 \text{ MeV}\)) of \(7.6 \times 10^{16} \text{n/(m}^2\text{s})\) \([17]\). Samples for research were prepared from the walls of irradiated ampoule. The samples cutting scheme is shown in Figure 1.

![Figure 1](image-url)  
**Figure 1.** Scheme of samples cutting for research: \(1–3\) are samples for thermal desorption spectrometry, \(4\) are samples for electron microscopy.

To eliminate of cold-hardening caused by mechanical action, the samples were subjected to the standard preparation procedure by mechanical grinding and polishing and final electro polishing in a
solution of 20% HClO4 + 80% C2H5OH [18]. Samples for TEM were prepared by jet electrolytic thinning in the same acid solution.

The presence of helium in steel after neutron irradiation and the peculiarities of its release were determined by helium thermal desorption spectrometry (HTDS) during uniform by rate heating of 0.7 K/s in the temperature range of 300–1273 K.

The microstructure of the irradiated samples was studied in TEM JEOL JEM 2100.

Mechanical characteristics of irradiated samples were determined by tensile tests using the LR5K Plus universal testing machine at temperatures of 298, 723 and 1033 K. After mechanical tests, the nature of fracture was studied in Hitachi TM4000Plus scanning electron microscope.

3. Results and Discussion

Figure 2 shows the HTDS spectra irradiated steel. TEM image of steel after irradiation is presented on Figure 3.

![Figure 2. HTDS spectra of 18Cr10NiTi steel irradiated by neutrons in a helium atmosphere at a temperature of 1033 K.](image)

![Figure 3. Microstructure of 18Cr10NiTi steel irradiated by neutrons in a helium atmosphere at a temperature of 1033 K.](image)
As can be seen in the HTDS spectra (see Figure 2), there are three peaks of helium release of different intensities in the temperature range of 300–625 K, which indicates the existence of several discrete stages with differing mechanisms of gas release during heating [19, 20]. Helium is insoluble in metals [5], it has extremely high mobility (for example, the migration activation energy of helium atoms is only 0.08 eV in Ni [22]), therefore, it may be trapped and retained with a sufficiently high binding energy on imperfections in the crystal structure only (vacancies, pores, interfaces between the second phases and the matrix, dislocations, grain boundaries and twins, etc.). The low-temperature location of HTDS peaks indicates a very low binding energy of introduced helium atoms with those of traps on which they are remain in irradiated steel up to temperature of ≤625 K.

Table 1 shows the activation energies of helium release in the TDS peaks in Figure 2, calculated by the Reherd formula [23].

| Peak number | E<sub>a</sub>, eV |
|-------------|------------------|
| I           | 2.1 ± 0.2        |
| II          | 2.2 ± 0.2        |
| III         | 2.5 ± 0.3        |

Accompanying all HTDS spectra first peak is observed at temperature of 453 K with low activation energy of helium desorption of 2.1 eV only. It is most likely due to the release of helium concentrated at a very shallow depth near the surface or adsorbed by the sample surface, since the binding energy of helium atoms with other possible defects is much more higher in metals [24, 25]. It should be noted that this peak does not always manifests itself clearly on different spectra of HTDS (see Figure 2).

The second and third peaks were detected at temperatures of 503 and 593 K respectively. Peak temperatures are too low compared to the irradiation temperature (T<sub>irrad</sub> = 1033 K). Therefore, it is difficult to identify their nature, but some assumptions can be made based on a number of known facts.

1. Complexes of a He<sub>m</sub>V<sub>n</sub> type are formed (m is the number of He atoms, n is the number of vacancies in the complex) during low-temperature (room temperature) introduction of helium. This complexes are thermally stable in nickel up to ~ 923 K at m = n [24, 25]. The possibility formation of thermally more stable complicated complexes of the He<sub>m</sub>M<sub>k</sub>V<sub>n</sub> type (k is the number of atoms of the alloying element in the complex) with a dissociation temperature of about 1033 K is shown after alloying of nickel by “oversized” elements (M = Al or Ti) [26]. However, T<sub>irrad</sub> is 1033 K in the present experiment; therefore, the formation of any helium-vacancy complexes is impossible, and helium bubbles (gas-filled pores) form immediately during irradiation by two- or polyatomic nucleation of their [5].

2. Having low solubility and high mobility, introduced helium “searches” traps for yourself, the most probable of which can be dislocations (steel was in the stat of 10% CW) and grain boundaries in the homogeneous 18Cr10NiTi steel (particles concentration of primary TiC carbide is negligible [2]). As can be seen on Figure 3, this is confirmed by the presence of gas pores (the dose of 1 dpa is too small for void formation [2, 27]) along the boundaries of austenitic grains, at triple junction points of grains and in the places of structural defects (dislocations) accumulation: gas-filled pores / bubbles appear light on the bright-field TEM image and they look as dark objects with a light border on the dark-field STEM image (shown in the insert).

Thus, the appearance of peaks II and III can be attributed to release of helium along grain boundaries and along dislocations [5, 19, 28]. Intensity of helium desorption in peak II significantly exceeds the intensity of helium release at stages I and III. Probably, this is associated with the gas outlet from the volume to surface along grain boundaries in peak II [5, 19]. But clarification required in what form helium releases grain boundaries (atomic, in the form of bubbles, or something else).
The manifestation of the peak of helium desorption at a temperature of 593 K remains to be associated with the release of helium through dislocation tubes [5, 19, 28, 29].

Figure 4 presents the results of mechanical uniaxial tensile tests of 18Cr10NiTi steel irradiated by neutrons in helium atmosphere. Figure 4 presents the results of mechanical uniaxial tensile tests of 18Cr10NiTi steel irradiated by neutrons in helium atmosphere. As can be seen, decrease in the ultimate tensile strength ($\sigma_{UTS}$) and relative elongation ($\delta$) are observed with increasing of test temperature $T_{test}$. Compared to investigation at room temperature, $\sigma_{UTS}$ decreases by ~16% and value of $\delta$ by ~19% at $T_{test} = 723$ K. A further increase in $T_{test}$ to 1033 K leads to softening of steel by ~64%, and a decrease in ductility by 53%. However, the change in the yield strength ($\sigma_Y$) as a function of $T_{test}$ is not monotonic: $\sigma_Y$ of steel increases by 25% with an increase in $T_{test}$ from 298 to 723 K. On the contrary, the test at $T_{test} = T_{irrad} = 1033$ K reduces it by 56%. As was shown earlier [30], an increase in $\sigma_{0.2}$ at $T_{test} = 723$ K can be associated with structural changes occurring in this irradiated by neutrons steel at a given temperature.

![Figure 4](image)

**Figure 4.** The dependence of mechanical properties on test temperature of 18Cr10NiTi steel irradiated by neutrons in a helium atmosphere.

Such behavior of the mechanical properties of steel saturated by helium in the process of high temperature neutron irradiation may indicate the occurrence of HTRO typical for fcc materials in the presence of helium, and this confirms the role of helium in this phenomenon [1–3, 5, 9–12].

The results of fractographic analysis of steel after mechanical tests are presented in Figure 5. There are a lot of small and large pores forming a dimpled relief in the fracture of the sample tested at room temperature (see Figure 5a). The nature of the fracture is viscous, like a “cup-cone” type. Fracture character is changed to brittle-ductile with increasing test temperature to 723 K (see Figure 5b). Destruction occurs predominantly along the grain boundaries, as evidenced by the presence of “tearing” formed at the site of carbide inclusions located along grain boundaries. With increasing $T_{isp}$ to 1033 to the fracture surface is a kink with a plurality of flat or slightly concave surfaces), and there are no obvious signs of plastic deformation (see. Figure 5c). There are cracks propagated by a brittle-viscous mechanism with a predominance of a brittle component. It should be noted that such a mechanism characteristic for materials destroyed at low temperatures [31].

And finally, you should mention another aspect relating to the mechanism of HTRE. Steel 18Cr10NiTi was entire period of irradiation in the reactor for 3280 h at $T_{irrad} = 1033$ K. This temperature is close to the temperature of maximum rate of high-chromium carbide of $M_{23}C_6$ type precipitation in austenitic steel [2]. However, we did not found significant traces of carbide inclusions, except for primary carbides / carbonitrides Ti (C, N), either in TEM studies of the microstructure or in fractographic studies of samples fractures. Titanium is added to steel in an amount of $C_Ti \geq 5\cdot(C_C - 0.03\%)$ ($C_C$ is carbon concentration in percents) in order to prevent intercrystalline corrosion by binding all carbon in TiC carbide so that chrome doesn't get carbon for the formation of $M_{23}C_6$ carbide along the grain boundaries. Thus, the effect of the second phases precipitates on the HTRE including grain boundary phase particles as suggested by the authors of several studies [1–3, 5, 9–12], is
excluded and the decrease in ductility during high-temperature tensile tests was due to the presence of helium precisely.

![Fractograms of breaking places of 18Cr10NiTi steel after mechanical testing at temperatures of 298 (a), 723 (b) and 1033 K (c).](image)

Figure 5.

4. Conclusions

The following conclusions can be made based on the results of studies influence of helium introduced into 18Cr10NiTi steel by an unusual method of ballistic recoil during the bombardment of a gas-filled balloon by fast neutrons.

1. At high temperature (1033 K) introducing of helium, it is trapped predominantly on structural defects with a low formation energy – grain boundaries and dislocations.

2. Helium release from irradiated steel occurs at very low temperatures with low activation energies during uniform postradiation heatings. This indicates a low binding energy of helium with these traps at high temperature.

3. The dependence of mechanical properties on test temperature of steel irradiated in a helium environment shows the manifestation of a high-temperature decrease in the ductility of steel in the presence of injected helium.

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