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Technical Note

Radiation damage to Ni-based alloys in Wolsong CANDU reactor environments

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Radiation damage due to neutrons has been calculated in Ni-based alloys in Wolsong CANDU reactor environments. Two damage parameters are considered: displacement damage, and transmutation gas production. We used the SPECTER and SRIM computer codes in quantifying radiation damage. In addition, damage caused by Ni two-step reactions was considered. Estimations were made for the annulus spacers in a CANDU reactor that are located axially along a fuel channel and made of Inconel X-750. The calculation results indicate that the transmutation gas production from the Ni two-step reactions is predominant as the effective full power year increases. The displacement damage due to recoil atoms produced from Ni two-step reactions accounts for over 30% out of the total displacement damage.

1. Introduction

Ni-alloys are employed widely in many industries since they have good mechanical and creep properties at high temperature, and superb corrosion resistance. However, they are not used extensively in nuclear reactors because of the undesirable nuclear properties of Ni element, including high absorption of thermal neutrons, and subsequently the radioactive activation of transmutation products [1]. In the cores of CANDU reactors, Ni-alloys are used as annulus spacers in fuel channel assemblies and as sheathing cables in flux detector assemblies. Garter spring-type spacers, made of Inconel X-750, are installed in the CANDU fuel channel to separate each pressure tube from a concentric calandria tube and to maintain a gap between the cold moderator (approximately 70 °C) and the relatively hot pressure tube (approximately 300 °C). Because the horizontal pressure tubes are likely to sag during service, four spacers per each pressure tube are arranged axially to prevent potential contact [2]. A key role of the spacers is to ensure that such sagging does not result in a pressure tube contacting the cooler calandria tube, which could lead to hydride formation at the contacted region followed by the cracking problem.

Recently, ex-service spacers have been found to have become embrittled after neutron irradiation in CANDU reactors [3,4]. Post-irradiation test of Inconel X-750 spacers exhibits lower tensile strength and significant intergranular failure [5]. Transmission electron microscopy (TEM) observation has shown the high density of nano-sized helium bubbles within the matrix and along the grain boundaries. Although the reason for such embrittlement is being investigated, the production of transmutation He gas is considered to be a major source of materials degradation. Because Inconel X-750 spacers are subjected to high neutron exposures during reactor operation, transmutation gas atoms continue to accumulate as a result of various neutron reactions, of which Ni two-step reactions are crucial in producing transmutation gas [1,6]. Also recoil atoms generated by the Ni two-step reactions aggravate displacement damage to materials. The amount of radiation damage in materials depends primarily on fast neutron irradiation that can lead to higher probability of atomic displacements and neutron transmutation reactions. Further it is noteworthy that thermal neutron irradiation brings about Ni two-step reactions.

In this study, we estimated the amount of radiation damage to X-750 spacers installed in the CANDU reactor theoretically. The damage parameters of interest include transmutation gas production and atomic displacement by neutron reactions. The neutron spectra for Wolsong Unit 2, a CANDU-type reactor in Korea, were

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employed for damage calculation. Using the neutron spectra near spacers, we calculated the production of transmutation gases of H and He by considering both the gas production reactions such as (n,p) and (n,α), and the Ni two-step reactions. In addition, displacement damage due to the recoil atoms of Ni two-step reactions, and the collision reactions between incident neutrons and lattice atoms, were estimated. Currently, a pressure tube material surveillance-program is underway for Wolsong Unit 2, which is a mandatory program under CSA N285.4 Section 12 [7]. Two pressure tubes were removed for inspection, followed by a series of standard material tests in the laboratory. In removing the two pressure tubes, eight X-750 spacers can be obtained as byproducts. Although the surveillance program of the removed spacers is not yet determined, those spacers are supposed to be stored for future use. The parameters of radiation damage to the X-750 spacers will be useful to predict their integrity through the modeling of irradiation embrittlement and creep. In addition, the calculation results will be helpful in analyzing the test data of the removed spacers when undertaking a spacer surveillance program.

2. Method of calculation

In this section, the calculation procedure is presented for quantifying the radiation damage to X-750 spacers in the CANDU core environments. First, we derive the neutron spectra for the spacer locations in Wolsong Unit 2 using the neutron transport code and prepare the latest neutron cross section data for the Ni two-step reactions. Subsequently, we describe the detailed methods for estimating the atomic displacement damage and the production of transmutation gas.

2.1. Neutron spectra and neutron cross section

Radiation damage to structural materials in nuclear reactors is caused by interactions between neutrons and lattice atoms. The fundamental factor affecting the amount of radiation damage is the neutron spectra at the location of interest. Hence, we calculated the neutron spectra for all fuel channel components in the Wolsong Unit 2 reactor using the WIMS-AECL code [8]. The average neutron spectrum for the whole reactor core and the neutron spectra for two X-750 spacers are shown in Fig. 1. Out of the two removed pressure tubes, one was designated as O14 and the other as Q11, both of which were located near the core center. The locations of the pressure tubes O14 and Q11 are shown in Fig. 2, which is an end view of CANDU reactor core. The inner diameter of the Calandria tube is approximately 6.75 m. The first spacer in the O14 pressure tube (O14 #1) is referred to as the outlet, while the second spacer in the Q11 pressure tube (Q11 #2) is close to the inlet. The spectrum is composed of 33 neutron groups with the slowest neutrons with energies ranging from 2.0×10^{-4} to 1.0×10^{-2} eV.

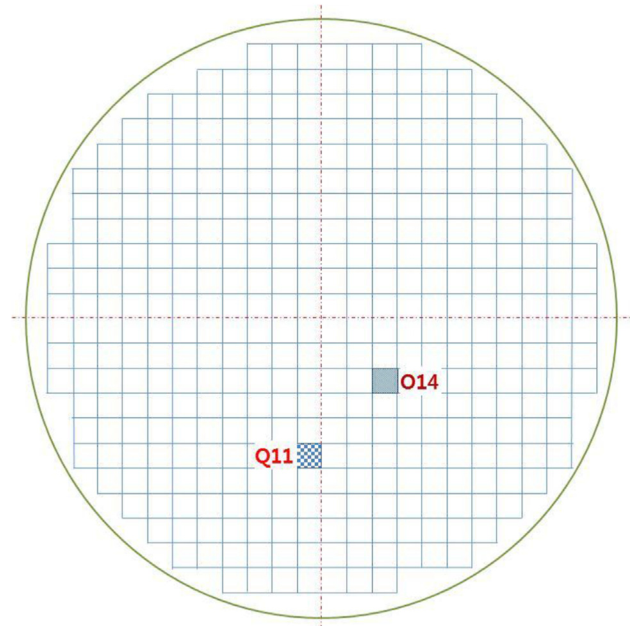

Fig. 2. End view of reactor core showing the locations of the pressure tubes O14 and Q11. (Calandria diameter ~ 6.75 m).

In most metals, transmutation gases are produced primarily by the absorption reactions with fast neutrons, such as (n,p) and (n,α) reactions. Further, Ni exhibits unusual nuclear properties with respect to interactions with thermal neutrons, thus leading to a higher production of transmutation gases. Both He and H can be produced by thermal neutron reactions with Ni via two-step reactions sequentially, such as ^{58}Ni(n,γ)^{59}Ni(n,α)^{56}Fe and ^{58}Ni(n,γ)^{59}Ni(n,p)^{59}Co, respectively. The amount of transmutation gas production depends on the neutron cross sections and neutron flux. The cross section data, which are necessary to estimate the gas production from Ni two-step reactions, were obtained from the latest ENDF/B-VII library [9]. Fig. 3 illustrates the relevant cross sections that were group-averaged based on the neutron energy group shown in Fig. 1. As shown in Fig. 3, we can expect that the

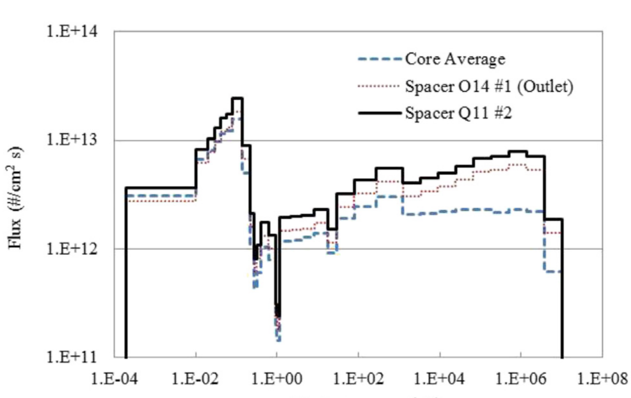

Fig. 1. Neutron spectra for the core and two spacer locations in Wolsong Unit 2 reactor.

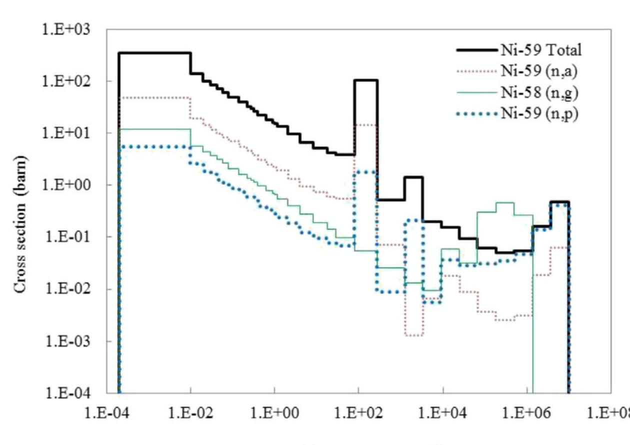

Fig. 3. Group-averaged neutron cross sections for ^{59}Ni(n,p), ^{58}Ni(n,γ), ^{59}Ni(n,α) and ^{59}Ni total absorption reaction as a function of neutron energy.

thermal neutrons are critical in the gas production from the Ni two-step reactions.

2.2. Transmutation gas production

The gas atoms of H and He can be produced by neutron interactions with constitutive elements in the metal. The production of He and its aggregation into bubbles causes metal embrittlement that cannot be eliminated by high-temperature annealing. On the other hand, the effect of H produced by neutron reactions on embrittlement is known to be minor because H atoms diffuse through metals rapidly and may lead to an escape from the component [5,6]. Although the measured H levels are close to the background [3] and there is no clear evidence of H trapping within bubbles, the estimation of H production is necessary to understand radiation damage mechanisms. The transmutations that produce such gas atoms can be divided into the following reactions: those that occur primarily in a fast neutron flux, and those that require a thermal neutron flux. The fast neutron flux causes direct (n,p) and (n,α) reactions in most component of the metal. Generally, transmutation reactions in the metals and the light impurity elements are of the threshold type, thus implying that the cross section is zero for all energy below a certain value. In estimating the gas production through direct neutron reactions, we can use the SPECTER computer code [10] that also allows us to obtain various radiation damage parameters including atomic displacements, recoil atom spectra, and spectrum-averaged cross sections. This code is compact and simple to use, and only the neutron spectrum is required as the input. Although we can readily obtain the amount of transmutation gas for the given neutron spectra from the SPECTER code, this code does consider the radiation effects of the Ni two-step reactions.

He atoms can also be produced by neutron reactions with Ni via the following sequential two-step reactions [11,12]:

$$^{58}\mathrm{Ni} + \mathrm{n} \rightarrow {}^{59}\mathrm{Ni} + \gamma$$

$$^{59}\mathrm{Ni} + \mathrm{n} \rightarrow {}^{56}\mathrm{Fe} + \alpha \tag{1}$$

The production of He atoms from the reactions (1) above is expressed as follows:

$$\frac{N_{He}(t)}{N_{Ni-58}} = \frac{\sigma_\alpha}{\sigma_T} - \frac{\sigma_\alpha \cdot \exp(-\sigma_\gamma \varphi_T\, t)}{\sigma_T - \sigma_\gamma} + \frac{\sigma_\alpha \sigma_\gamma \cdot \exp(-\sigma_T \varphi_T\, t)}{\sigma_T \cdot (\sigma_T - \sigma_\gamma)} \tag{2}$$

where N_{He} is the number of He atoms produced, N_{Ni-58} the initial number of ^{58}Ni atoms contained in metals, ϕ_T the neutron flux, t the irradiation time; σ_α, σ_T, and σ_γ are the nuclear cross section for ^{59}Ni(n,α), ^{59}Ni total absorption, and ^{58}Ni(n,γ) reaction, respectively.

H atoms can be produced by two methods: direct (n,p), and Ni two-step reactions, which are similar to the He production process [13]. The latter reactions can be expressed as follows:

$$^{58}\mathrm{Ni} + \mathrm{n} \rightarrow {}^{59}\mathrm{Ni} + \gamma$$

$$^{59}\mathrm{Ni} + \mathrm{n} \rightarrow {}^{59}\mathrm{Co} + \mathrm{p} \tag{3}$$

The production of H atoms from the reactions (3) above is given such as:

$$\frac{N_{H}(t)}{N_{Ni-58}} = \frac{\sigma_p}{\sigma_T} - \frac{\sigma_p \cdot \exp(-\sigma_\gamma \varphi_T\, t)}{\sigma_T - \sigma_\gamma} + \frac{\sigma_p \sigma_\gamma \cdot \exp(-\sigma_T \varphi_T\, t)}{\sigma_T \cdot (\sigma_T - \sigma_\gamma)} \tag{4}$$

where N_H is the number of H atoms produced and σ_p is the nuclear cross section for ^{59}Ni(n,p) reaction. In estimating the transmutation gas production, we use the SPECTER code, and Eqs. (2) and (4), including the neutron spectra and cross sections shown in Figs. 1 and 3, respectively.

2.3. Atomic displacement damage

Displacement damage to materials begins with the creation of primary knock-on atoms (PKAs) from the neutron interactions with lattice atoms. The PKAs are generated with a certain amount of energy that can cause multiple atomic displacements as they dissipate energy within a material. The PKA energy distribution is determined by the sum of recoils from various neutron reactions, including scattering and absorption reactions. We can obtain the PKA energy spectrum of elements from the SPECTER code [10]. Since the displacement cross sections are embedded in the SPECTER libraries, the displacement damage in units of displacement per atom (dpa) is readily obtained for a given neutron flux. It is well known that fast neutrons contribute significantly to atomic displacement damage primarily by elastic/inelastic scattering reactions. Accordingly, the estimated dpa values are proportional to the fast neutron fluence.

The direct contribution of thermal neutrons to displacement damage due to collisions is not significant. There are, however, additional displacements generated by the particles from the emission reactions, including photons, electrons, protons, neutrons, alpha particles, as well as recoil atoms. In the Ni two-step reactions, the recoil atoms and transmutation elements can affect the displacement damage. The ^{59}Ni(n,α) and ^{59}Ni(n,p) reactions, shown in Eqs. (1) and (3), respectively, generate both charged particles and heavy recoils that have kinetic energy. At thermal neutron energies, the ^{59}Ni(n,α) reaction produces 4.757 MeV alpha and 0.340 MeV ^{56}Fe atom whereas the ^{59}Ni(n,p) reaction produces 1.824 MeV proton and 0.031 MeV ^{59}Co atom. These recoils create many atomic displacements over their stopping range. A recoil atom loses energy in both electronic and nuclear collisions as it slows down and comes to rest in a material. Only the latter process causes lattice disorder around the recoil atom track and is responsible for displacement damage. The displacement damage due to the recoil atoms was calculated using the NRT damage energy model [14,15] and the SRIM code [16,17]. From the SRIM code calculation, we derived the fraction of recoil energy available for damage production and determined the damage energy. Table 1 displays the calculated values of the damage energy relevant to atomic displacement by four recoils atoms incident on Ni-Cr-Fe model alloys, which are extracted from the SRIM code calculations.

3. Calculation of radiation damage to X-750 spacers

The neutron spectra in the reactor cores and in the peripheral regions change significantly depending on location, which has a strong effect on the radiation damage. X-750 spacers are snug-fit on a pressure tube in a reactor core; they are separated axially from each other by approximately one meter. Depending on the axial position of the spacers, the total neutron flux is different whereas

Table 1
Amount of damage energy available for atomic displacements and projected ranges of four recoil atoms (H, He, Fe and Co) incident on Ni(70)-Cr(20)-Fe(10) model alloys. Results from SRIM code calculations.

	H	Co	He	Fe
Recoil energy (keV)	1824	31	4757	340
% energy loss to phonons by recoils (from SRIM)	0.1	72.06	0.25	56.17
Damage energy (keV)	**1.8**	**22.3**	**11.9**	**191**
Projected range (μm)	15.6	0.011	8.83	0.11

Table 2
Chemical composition in wt.% of Inconel X-750.

Element	Cr	Fe	Nb	Co	Mn	Cu	Al	Ti	Si	C	Ni
w/o	16.00	8.00	1.00	1.00	1.00	0.50	0.80	2.50	0.50	0.08	68.62

the shape of the energy spectrum for individual spacers is identical. Although it is possible to evaluate the radiation damage for all spacers installed in Wolsong Unit 2, we focused on two spacers including the first spacer of the O14 pressure tube (O14 #1) and the second spacer of the Q11 tube (Q11 #2). Among the eight spacers removed from the reactor, the O14 #1 and Q11 #2 spacers correspond to the lowest and highest neutron flux level, respectively.

The chemical composition of Inconel X-750 is listed in Table 2. In addition to Ni, all elements are taken into account in estimating the radiation damage.

3.1. Transmutation gas production

The calculated He production as a function of operation time for two spacer locations is shown in Fig. 4, where two neutron spectrum data for spacers O14 #1 and Q11 #2 are applied. The neutron spectra for each spacer are shown in Fig. 1. The He generation due to Ni two-step reactions is estimated using Eq. (2) and the relevant cross sections, and the He generation by direct (n,α) reactions is obtained from the SPECTER calculation. The He production by Ni two-step reactions begins to exceed that by the direct reactions after only several days of operation. It is clear that the He production by the Ni two-step reactions is dominant throughout the whole reactor operation. We can neglect the He generation by the direction reactions for a long service time. While the total He production for O14 #1 reaches approximately 1000 appm after 13 years of operation, about 10 years of operation is required to reach 1000 appm of He for the Q11 #2 spacer. In alloys containing Ni under thermal neutron irradiation, Ni two-step reactions play a dominant role in He production.

The H production for two spacer locations is plotted in Fig. 5. Unlike He production, H production from direct (n,p) reactions is

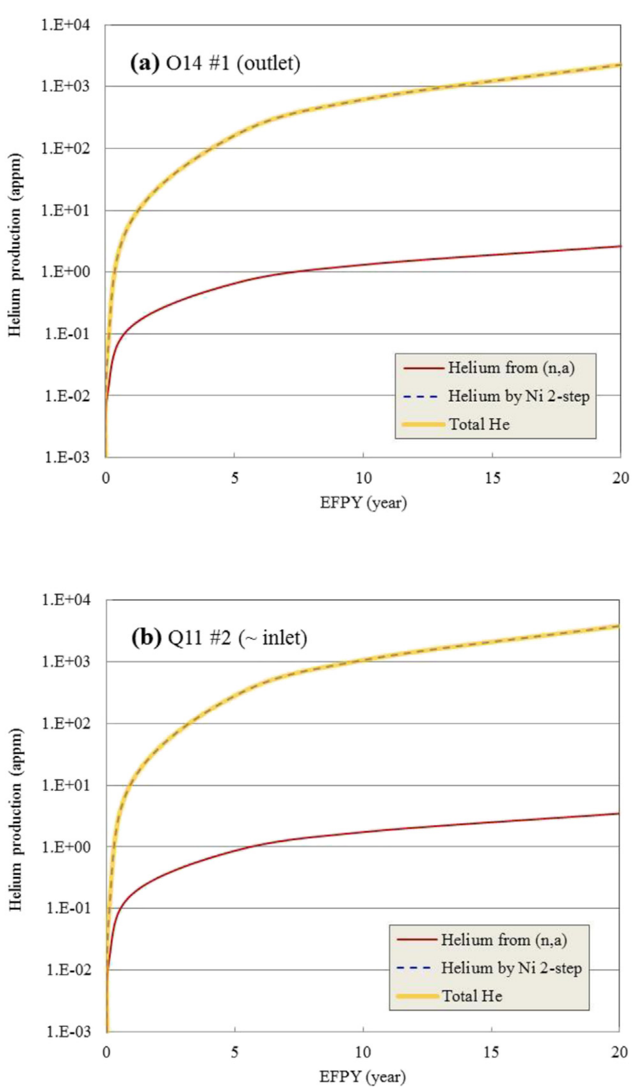

Fig. 4. Helium production for X-750 spacers in the core region of a CANDU reactor. (a) Spacer O14 #1 and (b) Spacer Q11 #2.

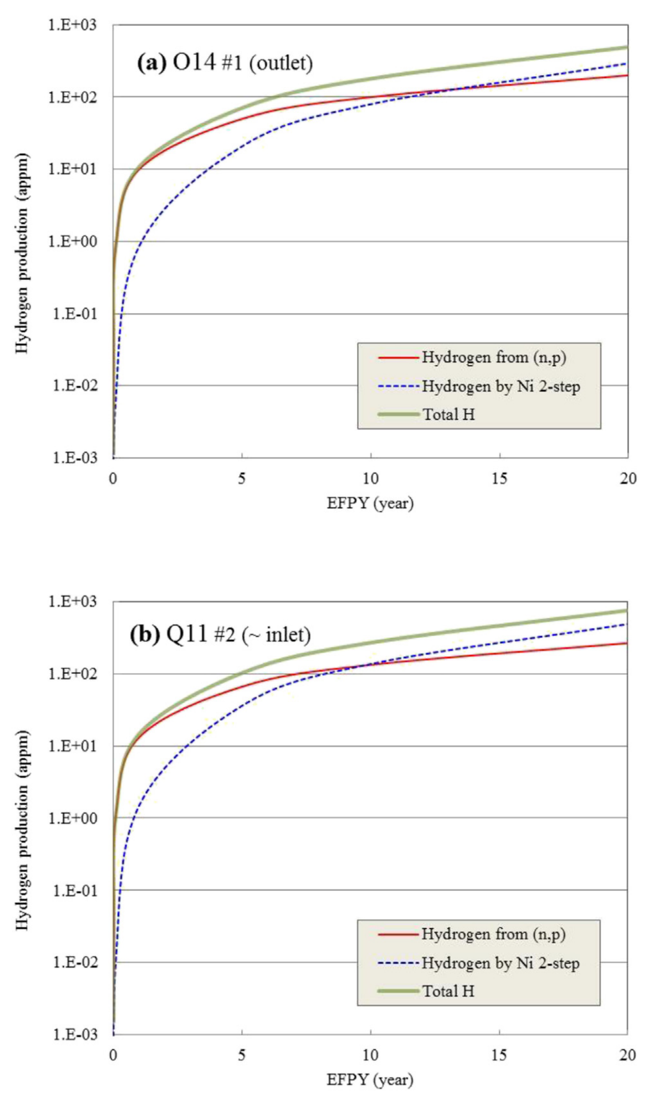

Fig. 5. Hydrogen production for X-750 spacers in the core region of a CANDU reactor. (a) Spacer O14 #1 and (b) Spacer Q11 #2.

predominant over the Ni two-step reactions in the initial period of operation. However, crossover takes place at about 10 years of service. In estimating the amount of H production, both the direct (n,p) and Ni two-step reactions must be considered.

3.2. Atomic displacement damage

We considered two effects of atomic displacement. One is the SPECTER code calculation that yields the dpa values for the given neutron spectra. This displacement is determined primarily by the fast neutron flux. As another source of atomic displacement damage, the damage due to recoil atoms from the Ni two-step reactions is considered. In the Ni-two step reactions, a one-to-one correspondence exists between the gas (H and He) production and the generation of recoil atoms. Based on this relation, we derived the helium-to-dpa and hydrogen-to-dpa ratios by combining the SRIM results [18] and the NRT model of damage energy [14]. The calculation results indicated that the dpa caused by $^{59}Ni(n,\alpha)$ reaction equals 1 dpa for every 492 appm of He. Similarly, the dpa caused by $^{59}Ni(n,p)$ reaction equals 1 dpa for every 4149 appm of H. The total displacement damage for two spacers were calculated, as shown in Fig. 6. While the displacement damage due to direct neutron reactions are linearly proportional to the reactor operation time, the damage by recoils from the Ni two-step reactions increases non-linearly. For 20 years of operation, the displacement damage by the Ni two-step reactions accounts for over 30% of the total damage; this must be considered in investigating radiation damage correctly.

4. Discussion

We herein emphasized the spectral aspects of Inconel X-750 alloys for the technological application to CANDU core components. The program of pressure tube material surveillance in Wolsong Unit 2 is currently in progress, in which a series of standard tests with Zr-alloys are to be conducted. Although specific guidelines on the treatment of removed X-750 spacers are not established, eight spacers are supposed to be stored in the hot cell to prepare for the integrity test. In Fig. 7, the calculated radiation damage, including the He & H production and dpa values, are displayed for eight spacers from the pressure tubes of O14 and Q11 at the time of 20-year operation. The difference in gas production among the spacers is slight, and the amount ranges from 2200 to 3800 appm of He and from 490 to 1020 appm of H; this is similar to the case of atomic displacement damage. When considering the spacer integrity test in the future, the calculated radiation damage is of importance to the exact evaluation.

Among the neutron transmutation gases, He is known to affect the microstructures and mechanical properties of alloys used in

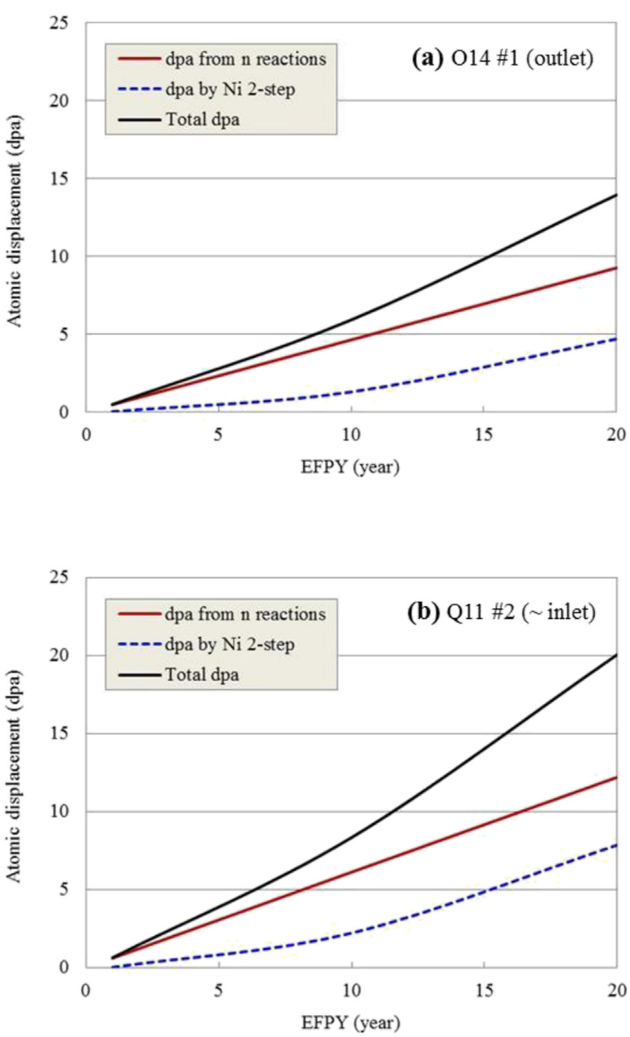

Fig. 6. Calculated atomic displacement damage for X-750 spacers in the core region of a CANDU reactor, (a) Spacer O14 #1 and (b) Spacer Q11 #2.

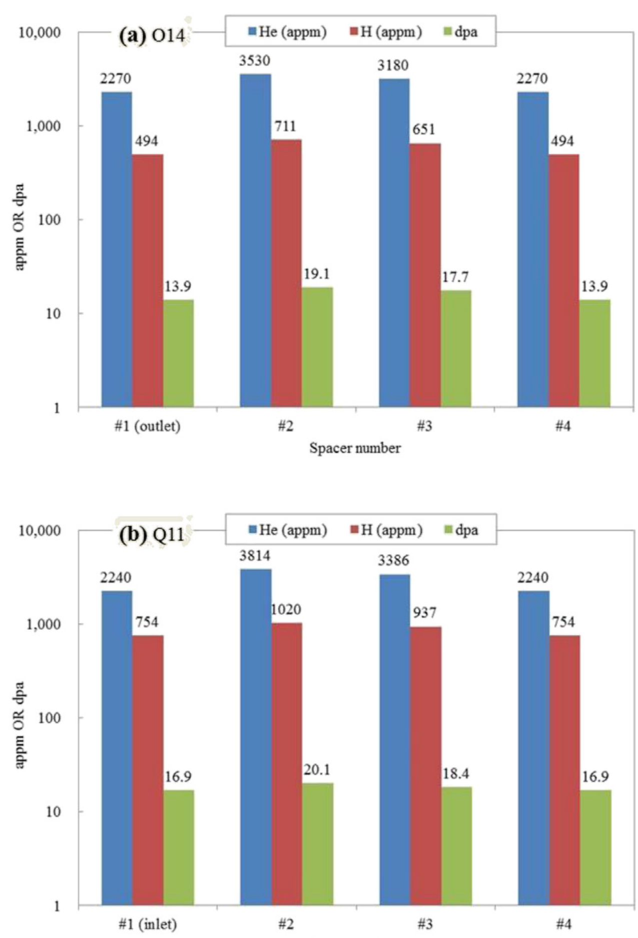

Fig. 7. Summary of radiation damage to X-750 spacers, including He and H production, and atomic displacement, at the time of 20-year operation. (a) O14 and (b) Q11 pressure tube.

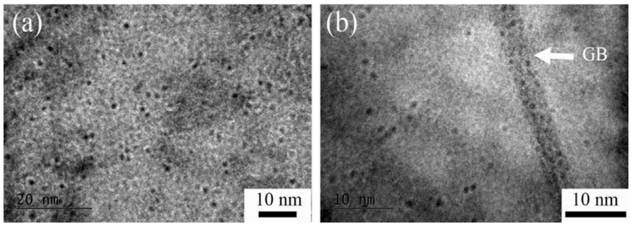

Fig. 8. TEM micrographs in the over-focused condition showing small-sized He bubbles (a) in the matrix and (b) near a grain boundary after ion implantation of 1000 appm He at 300 °C (Black dots: He bubbles, GB: grain boundary).

nuclear systems. High He concentrations in the alloys enhance radiation hardening and loss of ductility, and might lead to embrittlement problems. In order to understand the embrittlement phenomena of He-containing alloys, it is important to estimate the amount of gas production and displacement damage for the given neutron spectra, which are the primary subjects of this study. One of the experimental findings regarding the high-temperature embrittlement of He-containing metals reported that the embrittlement effect is noticeable at He concentrations of a few appm, and that its effect appears to saturate above He contents of approximately 10 appm [18]. Most of the He-containing samples were found to fail by intergranular fracture in contrast to He-free samples that break transgranularly. The analytical microscopy of X-750 spacers in the CANDU reactor showed that He atoms are present in the form of nano-sized bubbles within the grain boundaries and in the matrix [6]. He bubbles at the grain boundaries may be responsible for the intergranular fracture. The presence of He bubbles in ex-service X-750 spacers is well recognized by the TEM observation. Furthermore, He atoms were implanted into X-750 specimens at 300 °C using the α-particle beam to investigate the tendency to form He bubbles. The α-particle energy was varied between 1.4 and 2.8 MeV so that uniform distributions of 1000 appm He were implanted into the specimens of 2–5 μm depth. The calculated displacement damage from the SRIM code was approximately 0.1 dpa in the implanted region. The microstructure of Inconel X-750 after He-implantation was investigated by using TEM in the over-focus bright field imaging conditions. He bubbles were observed both in the matrix and in the grain boundary, which are shown in Fig. 8 (a) and (b), respectively. The density of bubbles is of the order of 10^{23} m^{-3} and their average size is ~1 nm in diameter. It is highly probable that He atoms tend to form bubbles in Inconel X-750 under He-implantation, as well as neutron irradiation. Griffiths *et. al* [1]. measured the level of He production from the X-750 spacers removed from the CANDU reactor to validate the calculations, which are described herein. The He concentration, as calculated from the CANDU spectrum and neutron cross sections, is in good agreement with the He contents measured in ex-service X-750 spacers. The calculated amount of He production for X-750 spacers in the Wolsong CANDU reactor is in the order of thousand appms, which is sufficient to cause intergranular failure.

In addition to X-750 spacers in the CANDU reactors [3–5], embrittlement was also found in the thimble tubes in pressurized-water reactors that are made from 316 stainless steels [19]. The tubes were irradiated to the range of 33–70 dpa at temperature of 290–315 °C. The TEM observation exhibited high-density Frank loops and nano-sized cavities at the grain boundaries, as well as within the matrix. Although the exact identity of cavities was not examined and no gas measurements were performed, they are thought to be gas bubbles containing high levels of He. It is highly probable that the presence of gas bubbles at grain boundaries may enhance the susceptibility of irradiated materials to intergranular failure. In addition to He production, the atomic displacement damage can affect the embrittlement phenomenon. As a result of atomic displacement reactions, a number of point defects in the form of vacancies and interstitials are generated. The vacancies are crucial in the growth of He bubbles on the grain boundaries when the tensile stress is applied to the component. Hence, a detailed calculation of displacement damage was conducted in this study. With respect to alloys containing significant amount of Ni under highly thermal neutron environments, particular attention needs to be paid to the evolution of microstructures, which cannot be overlooked because of the transmutation gas production and atomic displacement damage.

5. Conclusions

In this study, we estimated the amount of radiation damage to X-750 spacers under neutron irradiation. For Ni-based alloys, thermal neutron irradiation is important because Ni two-step reactions significantly affect the production of transmutation gas production (H and He) and the atomic displacement damage. To estimate the radiation damage to spacers in the Wolsong CANDU reactor accurately, we calculated the neutron spectra at the particular locations and prepared the cross sections for the Ni two-step reactions from the latest ENDF libraries. The evaluation was performed for the spacers that were removed from the pressure tubes O14 and Q11 in Wolsong CANDU reactor. The calculated amount of He production (in the magnitude of thousands of appm) and displacement damage (tens of dpa) is sufficient to give rise to embrittlement of the spacers. The calculation results will be useful in evaluating the status of the removed spacers when performing their integrity tests and in establishing a predictive model for mechanical property changes in Ni-based alloys.

Declaration of interest

None.

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