Calculation of neutron damage to Ni-containing components in CANDU reactors

Junhyun Kwon[a*], Gyeong-Geun Lee[a], Hyung-Ha Jin[a], Donghwan Park[b]
[a]*Korea Atomic Energy Research Institute, 989-111 Daedeok-daero, Daejeon, 34057, KOREA*
[b]*Korea Hydro & Nuclear Power Co.- Central Research Institute, 70-1312 Yuseong-daero, Daejeon, 34101, KOREA*
[*]*Corresponding author: jhkwon@kaeri.re.kr*

1. Introduction

Ni-alloys are employed in the cores of CANDU reactors as fuel channel spacers, tensioning springs. Recently it has been found that spacers, made out of Inconel X-750, became brittle after a long period of neutron irradiation and the production of helium (He) gas is considered to be an underlying degradation mechanism [1]. Garter spring-type spacers are installed to separate each pressure tube from a concentric calandria tube and to maintain a gap between the cold moderator (about 70^{o}C) and the relatively hot pressure tube (about 300^{o}C). In operating CANDU reactors, four annulus spacers are located axially along a pressure tube by about one meter. Pressure tubes are likely to sag between spacers because of the axial separation between the spacers in the horizontal channels. A key role of the spacers is to ensure that such a sagging does not result in a pressure tube contacting the cooler calandria tube, which may lead to hydride formation at the contacted region and further the potential for cracking.

Although the effect of He on radiation-induced microstructure, such as swelling, in Ni-containing alloys has been studied, that was not a big issue in the light water reactors since their operating temperature is low and the Ni contents contained in the structural alloys are not high. Fuel channel spacers in CANDU reactors are, however, subjected to high neutron exposures for a long time and made of Inconel X-750. It is well-known that He atoms are readily generated as a result of Ni two-step reactions. In addition, the nuclear properties of Ni in the thermal neutron environment tend to aggravate damage of materials due to displacement and gas production. Here, we quantify the primary radiation damage to Inconel X-750 in the CANDU core environments. The damage parameters of our interest include displacement damage and the amount of gas production (helium and hydrogen). In particular, the effect of Ni two-step reaction on damage is taken into account in this work.

2. Neutron Damage Parameters

In this section several neutron damage parameters are described, which include helium & hydrogen production by the direct and the *indirect* neutron reactions, and atomic displacement by primary knock-on atoms (PKA) and *indirect* activation products. Here, *indirect* stands for Ni two-step reactions. Firstly, we calculated the neutron spectra the CANDU reactor. Fig. 1 shows two spectra for a core average and a specific spacer location. The spectrum is composed of 33 neutron energy groups and the spacer spectrum in Fig. 1 is experienced by the X-750 spacer which is close to the core center. All analysis for the neutron damage calculation will be performed on the basis of the spacer neutron spectra.

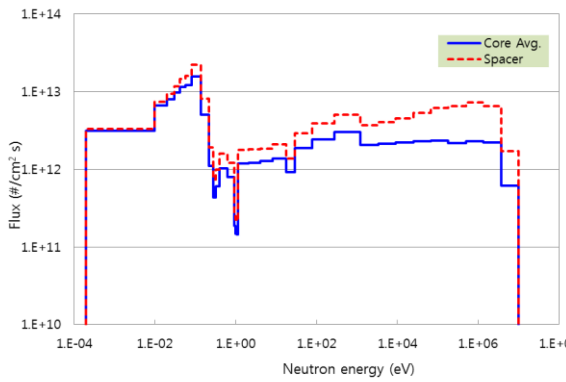

Fig. 1. Neutron spectra for an average CANDU channel power and X-750 spacer

2.1 Helium Production

He is known to be thermodynamically insoluble in metals and to form bubbles on the grain boundaries at high temperature (> 450^{o}C). These bubbles on the grain boundaries can lower the ductility of the metals. He is directly produced by fast neutron reaction with metal elements and by thermal neutron one with boron contained in metal as an impurity. The amount of He production by direct (n,α) reactions can be obtained from the SPECTER code calculation with an input of neutron spectra [2].

He can also be produced by neutron reaction with Ni via the following two-step reaction.

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

$$^{59}\mathrm{Ni} + \mathrm{n} \rightarrow {}^{56}\mathrm{Fe} + {}^{4}\mathrm{He} \qquad (1)$$

The production of He from the above reactions is given by [3]:

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

where N_{He} is the number of He atoms produced, $N_{Ni\text{-}58}$ is the initial number of ^{58}Ni atoms contained in X-750, ϕ_T

is the neutron flux; t is 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) reactions, respectively. The cross section data for the above three reactions, obtained from the latest ENDF/B-VII library, are shown in Fig. 2. Those values are collapsed for given neutron energy group.

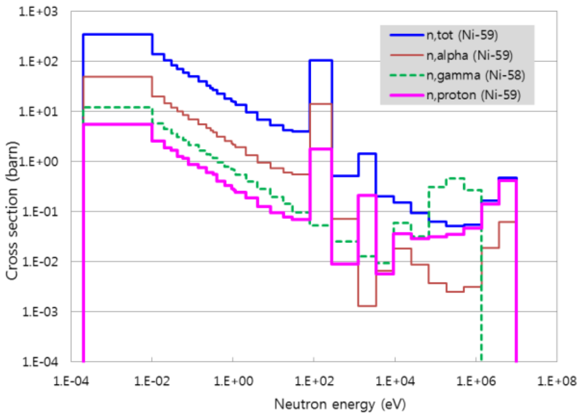

Fig. 2. Neutron reaction cross sections for ^{59}Ni total, ^{59}Ni(n,α), ^{58}Ni(n,γ) and ^{59}Ni(n,p) reactions as a function of neutron energy

2.2 Hydrogen Production

Like the similar process to the He production, hydrogen (H) can be produced in two ways, direct (n,p) and Ni two-step reactions. The latter reactions can be expressed in Eq. (3).

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

$$^{59}\text{Ni} + \text{n} \rightarrow {}^{59}\text{Co} + \text{H} \qquad (3)$$

The production of H from the above reactions is given such as [4]:

$$\frac{N_H(t)}{N_{Ni-58}} = \frac{\sigma_p}{\sigma_T} - \frac{\sigma_p \cdot \exp(-\sigma_\gamma \phi_T t)}{\sigma_T - \sigma_\gamma} + \frac{\sigma_p \sigma_\gamma \cdot \exp(-\sigma_T \phi_T t)}{\sigma_T \cdot (\sigma_T - \sigma_\gamma)} \qquad (4)$$

where N_H is the number of H atoms produced and σ_p is the nuclear cross section for ^{59}Ni(n,p) reaction. The group-averaged cross sections are shown in Fig. 2. Most procedures are almost the same as those for the He production calculation.

2.3 Atomic Displacement

Neutron damage to materials begins with the creation of PKAs from high-energy neutron-nuclear interactions. The PKA spectrum is determined by such factors as the incident neutron energy, the masses involved, and the angle between the incident neutron and the recoil direction. We can readily obtain the PKA spectrum of various elements for a given neutron spectrum from the SPECTER code calculation [2]. Accordingly, for a given neutron energy spectrum, the SPECTER simply calculates the damage parameters of displacement per atom (dpa). Dpa represents the calculated number of recoil atoms that are displaced from their lattice sites as a result of particle collision. The displacement damage due to direct nuclear reactions with elements, including scattering and absorption ones, can be readily estimated using the SPECTER.

The contribution of slow neutrons to displacement damage is not significant. However, the transmutation elements produced by Ni two-step reactions can have a great effect on the displacement damage. The ^{59}Ni(n,p) and (n,α) reactions, shown in Eqs. (1) and (3), generate both charged particles and heavy recoils which lead to displacement damage. At thermal neutron energies, the (n,α) reaction produces a 4.757 MeV alpha and 0.340 MeV ^{56}Fe atom whereas the (n,p) reaction produces 1.824 MeV proton and a 0.031 MeV ^{59}Co atom. The displacement damage due to these reactions was calculated using the Lindhard energy partition model. It was shown that the net dpa value caused by ^{59}Ni(n,α) equals 1 dpa for every 567 appm of He. Since these values are calculated on the basis of thermal neutron energy, the exact ones are variable depending on the given neutron spectrum.

3. Damage Calculation Results

The following calculation results are based on the neutron spectrum of the specific spacer shown in Fig. 1. The target material is an Inconel X-750 for the annulus spacer and its chemical composition is listed in Table 1.

Table 1. Major elements (w/o) in 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

3.1 Helium Production

The calculated He production in one gram of X-750 as a function of operation time is shown in Fig. 3. While He generation due to Ni two-step reactions is estimated using Eq. (2), the amount of He generation by direct (n,α) reactions is obtained from the SPECTER calculation. After fifteen days of operation, the He production by Ni two-step reactions exceeds that by direct reactions. It is clear that the He production is dominated by Ni two-step reactions throughout the whole reactor operation. The total He production reaches about 1,000 appm after 11 years of operation. In alloys containing Ni under thermal neutron irradiation, the source of He production should be taken into account even at low temperature.

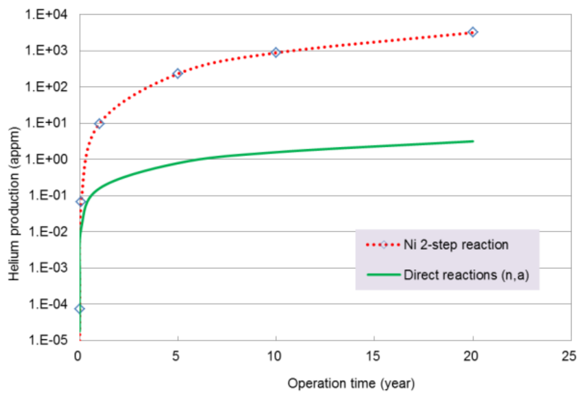

Fig. 3. Calculated He production for Inconel X-750 (1 gram) in the spacer of CANDU reactor

3.2 Hydrogen Production

The H production as a function of operation time is shown in Fig. 4. At the beginning of reactor operation, the H production is dominated by the direct (n,p) reactions. However, crossover takes place at about ten years of reactor operation.

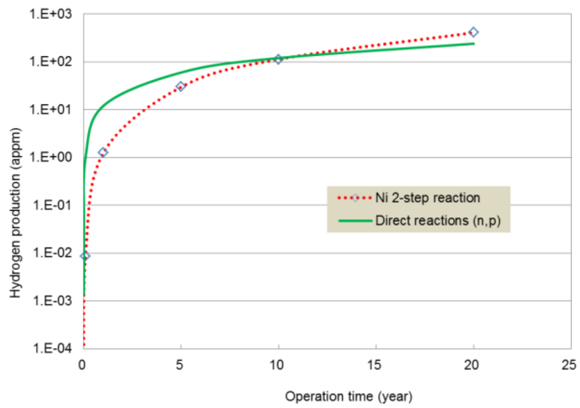

Fig. 4. Calculated H production for Inconel X-750 (1 gram) in the spacer of CANDU reactor

3.3 Atomic Displacement

We have calculated displacement damage produced by fast neutrons using the SPECTER code. In addition, the contribution of ^{59}Ni(n,α) reaction to damage was taken into account by considering the kinetic energy of transmutation product of ^{56}Fe and He particles. While the displacement damage due to fast neutrons was 11.1 dpa for twenty years of reactor operation, whereas the additional damage due to ^{59}Ni(n,α) reaction was 5.6 dpa.

4. Conclusions

In dealing with Ni-rich alloys such as Inconel X-750 under thermal neutron environment, we have to consider the effect of radiation damage due to Ni two-step reactions. In this work, three parameters of neutron damage were estimated, which include He and H gas production, and atomic displacement. The neutron spectrum of the CANDU reactor core were produced by the neutronics code and the latest nuclear data from ENDF/B-VII were employed for interpreting Ni two-step reactions. Calculations predict that high concentrations of He and H are to be expected in Inconel X-750 of an annulus spacer after neutron irradiation. In particular, the He production due to Ni two-step reactions is dominant in the early stage of reactor operation. Besides, the displacement damage due to transmutation product from Ni two-step reactions is increasingly significant, which is not negligible. Over 20 years operation, displacement damage by Ni two-step reaction reaches one-third of the total dpa value.

The above evaluation has been made without specific power history. Accordingly, the amount of radiation damage might be changed depending on the irradiation condition. This work implies that Ni two-step reaction would be considered for the evaluation tool of Ni-containing materials under long-term irradiation conditions. Further work is required with detailed power history and component geometry. However, the calculation results will provide basic information for predicting irradiation creep and He embrittlement of Inconel X-750 under thermal neutron conditions.

REFERENCES

[1] M. Griffiths, G.A. Bickel, S.A. Donohue, P. Feenstra, C.D. Judge, D. Poff, L. Walters, M.D. Wright, L. Greenwood, F.A. Garner, Degradation of Ni-alloy Components in CANDU Reactor Cores, 16th Int. Conf. on Environmental Degradation of Materials in Nuclear Power Systems – Water Reactors, Asheville, NC, USA, Aug. 11-15, 2013.

[2] L.R. Greenwood, K. Smither, SPECTER:Neutron Damage Calculations for Materials Irradiations, ANL/FPP/TM-197, 1985.

[3] L.R. Greenwood, A New Calculation of Thermal Neutron Damage and Helium Production in Nickel, J. Nucl. Mater. Vol.115, p.137, 1983.

[4] L.R. Greenwood, F.A. Garner, Hydrogen Generation Arising from the ^{59}Ni(n,p) reaction and its impact on fission-fusion correlations, J. Nucl. Mater. Vol. 233-237, p.1530, 1996.