Queen's University – Zhongwen Yao CRD

CRD Title: Aging of Inconel X-750 Spacer Material

Overview

Dr. Zhongwen Yao was awarded the NSERC/UNENE/ Collaborative Research and Development (CRD) Grant on June 1, 2013.

In modern CANDU® reactors, spacers are tight fitting springs that provide support to the pressure tube, separate it from the cold calandria tube (80°C) and prevent creep deformation of the pressure tube. It is essential that the spacers in fuel channels maintain their integrity throughout the fuel channel life, so the pressure tubes can be guaranteed not to come in contact with the calandria tubes, risking hydride blister formation and pressure tube rupture in which case the nuclear power plant would be accidentally shut down. The first generation of spacers were made of Zr-2.5Nb-0.5Cu and designed to fit loosely with pressure tube. However the relaxation of the spring due to irradiation led directly to the P2 G16 incident in 1983. As a result, the spacers in CANDU reactors were changed to a tight fitting design made of Inconel X-750. In principle, Inconel X-750 is a γ' Ni_3[Al, Ti] strengthened Ni based superalloy which possess excellent mechanical strength and good creep properties in addition to oxide and corrosion resistance at high temperatures. However, in recent years the effect of aging on the properties of Inconel X-750 spacers has been a growing concern for the CANDU industry. Often spacers from removed channels have been found to be broken. Mechanical tests on removed spacers have been conducted in CRL and results suggest that they may have become embrittled.

The main objective of this project is to develop an understanding of the embrittlement behaviour of the spacer, to address life management issues in existing operating reactors. The focus of this project is to examine the effects of displacement damage using energetic ions (as an analogue to fast neutron irradiation), combined with the effect of helium (via implantation carried out using tandem accelerators, to simulate the effect of helium produced by n-α reactions from Ni) on the properties of Inconel X-750: Ni (70%min), 13-17 Cr, 5-9 Fe, 2.25-2.75 Ti, 0.4-1.0 Al, 0.7-1.2 Nb+Ta. The short and long term results are directly applicable to understanding the effect of aging on the integrity of CANDU spacers. The techniques and analysis methods applied here to ion irradiated materials help characterize neutron irradiated materials being studied at the Chalk River Laboratory (CRL) in parallel where the work can be rather challenging in terms of the complexity in operation of radioactive materials. The following sections outline progress achieved during 2016-2017.

Program Results /Highlights & Advances in Knowledge

In this study, ion irradiation with helium pre-implantation is primarily employed in bulk materials. The investigation was mainly carried out at Montreal, Quebec. The figure 1 illustrates irradiation facility in University of Montreal, and nano-indentor in reactor materials testing lab (RMTL) of Queen's University. The samples were characterized using transmission electron microscope prior to irradiation. Figure 2 indicates the element distribution of X-750 raw material which implied the dual phase structure of this material - γ / γ' and they are in ordered crystal structure.

Fig. 1 A) An accelerator @ University of Montreal was used for irradiation and helium implantation of samples. B) A Nano-Indentation @ Queen's University was used for mechanical testing.

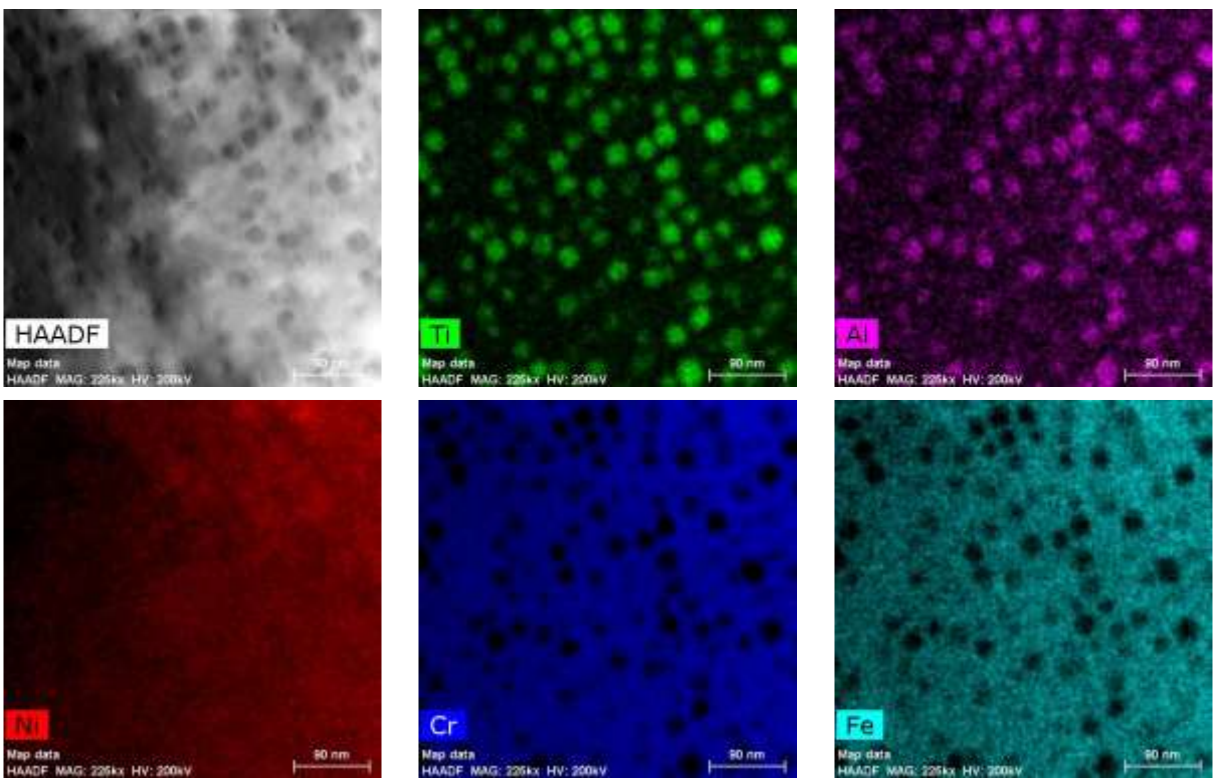

Fig. 2. ChemiSTEM mapping of X-750 prior to irradiation showing elements distribution within γ' precipitates.

1) **To characterize the microstructures of 40 MeV Ni ion irradiation induced defects in spacer X-750 alloys, with and without helium pre-implantation.**

The Ni ion irradiation is supposed to be closer simulation of fast neutron irradiation (~ 1 – 10 dpa), whereas the helium implantation (5000 appm helium) acts as transmutation production mainly from thermal neutron damages. The focus is given to the microstructure evolution of Inconel X-750 after irradiation using TEM characterization. The results indicated:

- In the sample irradiated at room temperature, the cavity density is lower than the sample irradiated at 400 °C, and the cavities average size is bigger (2±0.2 nm). The cavity size distribution diagrams in both helium pre-implanted samples are depicted in figure 3.
- The γ' superlattice reflections were found at different locations in the irradiated region as well as the un-irradiated layer in the He-Ni at 400 °C and Ni at 400 °C samples. In contrast, in the He-Ni at RT sample superlattice reflections disappeared in the irradiated region. Therefore, the γ' phase stayed ordered after helium implantation and subsequent Ni irradiation at 400 °C as well as single Ni ion irradiation at 400 °C; however, disordering of γ' happened under the condition of He-Ni at RT.
- A larger size and a higher density of Frank loops were obtained for the pre-implanted helium sample irradiated at 400 °C, compared to the sample irradiated at the same temperature by single Ni+ ions. Also, increasing the self-ion irradiation temperature led to an increase in the size and the density of the Frank loops.
- A large amount of small defects (mainly ~ 2nm stacking fault tetrahedra) were observed in all samples.

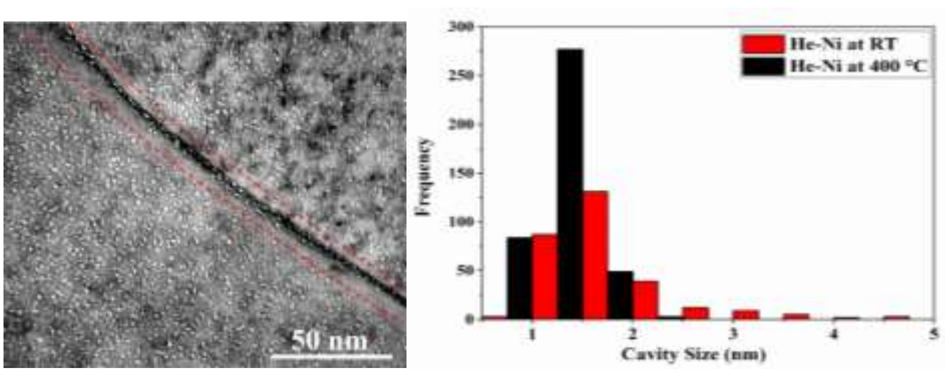

Fig. 3. Cavity size distribution in He-Ni at 400 °C and He-Ni at RT irradiated X-750.

2) **To measure the irradiation induced changes on mechanical properties of spacer X-750 alloys, using Nano-indentation.**

The measured nano-hardness profiles as a function of indentation depth are presented in Fig. 4. The effect of pre-helium implantation on indentation behaviour of X-750 alloy is clear in Fig. 4a. The results show:

1) The irradiation-induced hardening appeared for both He-Ni at 400 °C and Ni at 400 °C; however, the level of hardness enhancement is more in the pre-helium implanted sample compared to single Ni ion irradiated sample. Indeed, pre-helium implantation increased the hardening effect of Ni+ ion irradiation during the deformation of X-750 alloy. Furthermore, the influence of subsequent self-ion irradiation temperature on nano-hardness of X-750 alloy is presented in Fig. 4b. The hardness profiles depict that He implantation and subsequent self-ion irradiation at

400 °C made the material harder than the un-irradiated material. In contrast, changing the Ni+ ion irradiation temperature to room temperature (26 °C) resulted in a softening behaviour of the material.

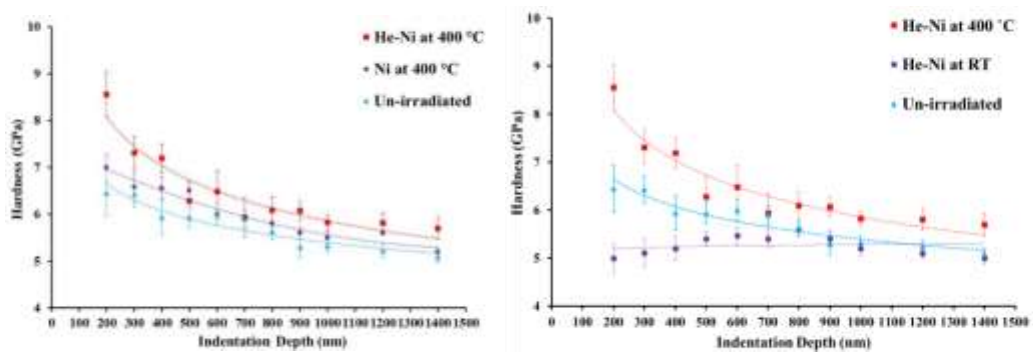

Fig. 4. Nano-hardness variations as a function of indentation depth, showing the effect of a) pre-helium implantation and b) Ni+ ion irradiation temperature

2) Three different hardening models were employed to calculate the barrier strength from different obstacles.

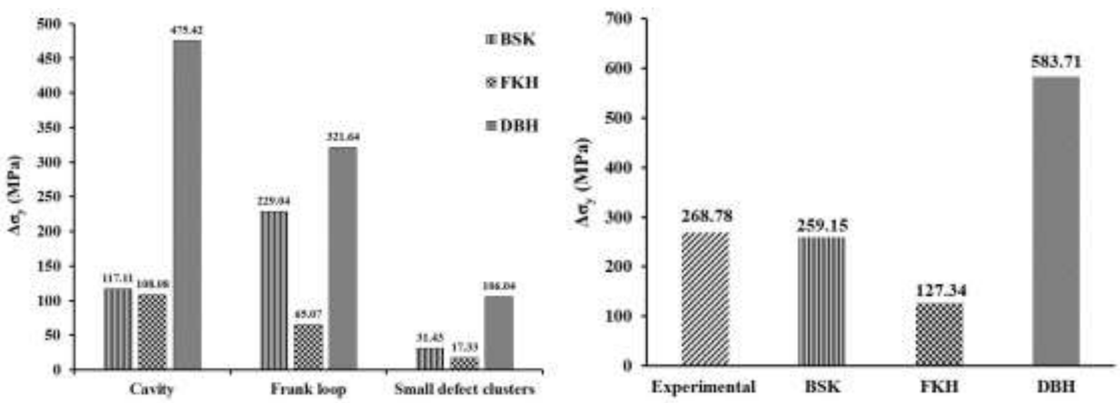

Fig. 5. The results of obstacle hardening models showing a) individual contribution of each type of defects to the irradiation-induced hardening and b) superposition of each model and comparing to experimental result.

The dispersed barrier hardening (DBH) model predicts the yield strength increase caused by arbitrary defects impeding dislocation motion. An alternative hardening model was proposed by Friedel–Kroupa–Hirsch (FKH) and describes material hardening due to the presence of circular dislocation loops. The third model is the Bacon-Kocks-Scattergood (BKS) model which describes the increase in yield strength when there is an infinite array of impenetrable spherical obstacles. The total increase in yield strength can be predicted using superposition principle to combine the independent defect hardening contributions according to this equation:

$$\Delta\sigma_T^n = \Delta\sigma_{Cavity}^n + \Delta\sigma_{Frank\ loop}^n + \Delta\sigma_{Small\ defect}^n$$

As figure 5 shows the fitting results from three models using the data obtained from experimental measurement, the best fit for the strength variation due to irradiation was accomplished using a superposition of the BKS model. According to the obtained contribution of each defect type to the irradiation hardening based on the BKS model, the large interstitial Frank loops have the most significant impact on the mechanical property changes, followed by cavities, which play a relatively strong role inducing irradiation hardening. Finally, it was found that smaller defects cause a minor hardening effect on the overall mechanical response of the irradiated X-750.

Overall, our progress on the project well followed the steps of original proposal.

Cases with Realized outcomes to Industry; Knowledge Transfer, Technology Application to Industry etc.; same as above

The CRD research is to assist the ongoing work in AECL, 'Fuel Channel Life Management / Microhardness of Inconel X-750'.

Research Facilities and Equipment Established Recently or in the Near Future also Existing ones

1. A computer cluster was built for calculating the damage morphology in alloys.
2. A sophisticated TEM lab is being built with in-situ heating, straining, 3D construction and low background measurement capabilities, considered as unique equipment in Canada.

Current HQP enrolled in the programs; number and type

The NSERC/UNENE CRD involved close collaboration with Prof. Mark Daymond of Queen's University, Dr. Colin Judge of the AECL-CRL in the areas of microstructure characterization and rate theory calculation of Ni superalloy.

Eight HQP are being trained by this CRD project. Namely, Ken Zhang (PhD, Queen's), Sali Di (PhD, Queen's), Iris Wang (MSc, Queen's), Pooyan Changizian (PhD, Queen's), Qingshan Dong (PhD, Queen's), Adam Brooks (UG, Queen's), Fengfeng Luo (PhD, Wuhan U), Yuanze Sun (MEng, Queen's).

HQP that had Graduated; number, type and *specify where employed*

Mr. Adam Brooks has been hired by AMEC Bruce Power.
Ms. Iris Wang has been hired as researcher in Beihang University in China.
Dr. Fengfeng Luo has been hired as staff scientist in Nanchang Institute of Applied Physics in China.

Publications / Journal Papers

Peer-reviewed Journal Paper

1. Changizian*, P., Lu*, C., & Yao, Z. (n.d.). Indentation behaviour of ion-irradiated X-750 Ni-based superalloy. Philosophical Magazine Letter, 97(3), 101–109, (2017).
2. Dong, Q., & Yao, Z.. Precipitate Stability in a Zr–2.5 Nb–0.5 Cu Alloy under Heavy Ion Irradiation, Metals, 7 (8), 287 – 301, (2017).
3. Dong, Q., Yu, H, Daymond M., & Yao, Z.. Study of microstructure and precipitates of a Zr-2.5 Nb-0.5 Cu CANDU spacer material, Journal of Nuclear Materials, 481, 153 – 163, (2016).
4. Changizian*, P., Zhang*, K. H., & Yao, Z. (n.d.). Effectof simultaneous helium implantation on the microstructure evolution of InconelX-750 superalloy during dual beam irradiation. Philosophical Magazine, 95(35), 3933–3949, (2016).
5. Yao Z, Zhang KH*, Zhou Z, Daymond MR, S Jublot-Leclerc, O Kaitasov, Materials Science and Technology, 1-5, (2017)

Conference Oral Presentation

1. Yao Z., (2016) 'Microstructural Characterization of Irradiated Inconel X-750 Superalloy Using Dual Beam (Ni+/He+) Irradiation at High Temperature', Invited Talk, 27th International Conference on Atomic Collisions in Solids (ICACS), Lanzhou, China.
2. Yao Z., (2016) 'Irradiated Inconel X-750 Superalloy by fast neutrons vs thermal neutrons', Invited Talk, 4th NuMat, Montpelier, France.
3. Changizian, P., & Yao, Z. (2017). Microstructure characterization of dual beam (Kr+2/He+) irradiated Inconel X-750 spacer material. Fuel Channel Seminar.
4. Yao Z. (2016). The ion irradiation of X750 at Queen's University, Ottawa, One day Ni alloy meeting.
5. Dong, Q., & Yao, Z. (2016). Effect of annealing and irradiation on the microstructure and phase stability of a potential CANDU Zr spacer. UNENE Annual Meeting.
6. Changizian, P., & Yao, Z. (2016). Mechanical properties of irradiated X-750 spacer material evaluated with nano-indentation methods. UNENE Annual Meeting.
7. Yu, H., Yao, Z., & Daymond, M. (2016). Effect of heavy ion irradiation on the microstructure of Zr-Excel alloy pressure tube. Fuel Channel Seminar.
8. Yao, Z., & Di, S. (2016). Component Dislocation Loop Nucleation in Zirconium under Irradiation: Atomic-level Simulations. Fuel Channel Seminar.
9. Yao, Z., & Zhang, K. (2016). Radiation Induced Degradation of CANDU Spacers. Fuel Channel Seminar.

Conference Poster Presentation

1. Changizian* P. & Yao, Z. (supervisor). Indentation behaviour of ion-irradiated X-750 Ni-based superalloy., 2017 CANDU Owner Group (COG) Fuel Channel Seminar, June, 2017. *Poster won 2nd Prize ($500).
2. Dong, Q., & Yao, Z. (supervisor). Effect of annealing and irradiation on the microstructure and phase stability of a potential CANDU Zr spacer., 2017 CANDU Owner Group (COG) Fuel Channel Seminar, June 2017.
3. Cong D., Yao,Z. and M.R. Daymond, 'The habit plane of⟨ a⟩-type dislocation loops in α-zirconium: an atomistic study', 2017 CANDU Owner Group (COG) Fuel Channel Seminar, June 2017.
4. Dong, Q., Yao, Z. Daymond M. 'Precipitate Stability in a Zr–2.5 Nb–0.5 Cu Alloy under Heavy Ion Irradiation', 2017 CANDU Owner Group (COG) Fuel Channel, Toronto, June, 2017.

Technical Reports

1. Zhang K., Yao Z., Heavy Ion Irradiation of Inconel X-750 Spacer Material, COG-14-1050 report.
2. Balogh L., Long F., Yao Z. and Daymond M., Quantifying irradiation defects in Zr alloys: a comparison between transmission electron microscopy and whole pattern diffraction line profile analysis. STP- Technical Paper 2017.

<u>Interactions /Consultations to Industry or Others</u>

This CRD program directly benefits the parallel study of neutron irradiated spacer program in Chalk River Lab. The university research involved strong collaboration with AECL–CRL, Kinectrics and Argonne National Lab.

Dr. Yao has refereed the scientific proposals for EMIR JANNUS in France. He refereed one funding proposal for the Sylvia Fedoruk Canadian Centre for Nuclear Innovation.

Dr. Yao also reviewed a number of manuscripts for Acta Materialia, Materials Characterization, Philosophical Magazine, Journal of Nuclear Materials, Journal of Alloys and Compounds, Journal of Materials Science, Journal of Microscopy, Computational Materials Science and CNL Nuclear Review,.