Heavy Ion Irradiation Effects in Zr Excel Alloy Pressure Tube Material

Y. Idrees[1], Z. Yao[1], M. Sattari[1], M.R. Daymond[1]
Idreesy@me.queensu.ca
1- Department of Mechanical and Materials Engineering, Queen's University, Kingston, ON, Canada, K7L3N6

Summary

Zirconium Excel alloy (Zr-3.5wt.%Sn-0.8%Nb-0.8%Mo) is the candidate material for pressure tubes in the Generation-IV CANDU® Super Critical Water-cooled Reactor (SCWR) design. Changes in microstructure induced by neutron irradiation are known to have important consequences on the in-reactor deformation behavior. The in-situ ion irradiation technique has been employed to elucidate the irradiation damage in dual phase Zr-excel alloy (~60% hcp alpha and ~40% bcc beta). 1 MeV Kr ion irradiation experiments were conducted at different temperatures ranging from 100°C-400°C. Damage microstructures have been characterized by Transmission Electron Microscopy in both the alpha and beta phases at different temperatures after a maximum dose of 10 dpa. Several new observations including irradiation induced omega (ω) phase precipitation have been reported. The ω/β orientation relationship was determined by the detailed analysis of selected area diffraction patterns. In-situ irradiation provided an opportunity to observe the nucleation and growth of basal plane c-component loops. It has been shown that under Kr ion irradiation the c-loops start to nucleate and grow above a threshold dose, as has been observed for neutron irradiation. Furthermore, the role of temperature, material composition and pre-irradiation microstructure has been discussed in detail.

1. Introduction

Zirconium-Excel alloy is a possible candidate for the pressure tube material for two of the fuel channel designs being considered for the CANDU-SCWR, a pressure-tube type supercritical water cooled reactor [1]. First is the High Efficiency Channel (HEC) and the second is Re-entrant Channel (REC). Excel (Zr - 3.5%Sn - 0.8%Nb - 0.8%Mo - 1130 ppm O) is a high strength, creep resistant zirconium alloy developed by AECL in the 1970s [1-3]. In the HEC design, the pressure tube would operate at ~80°C, whereas in re-entrant design the pressure tube would operate at a temperature of about 350°C to 400°C.

Radiation damage in components of nuclear reactors alters the mechanical properties, making these parts susceptible during service to hardening, loss of ductility, localised plastic deformation and plastic instability [4]. Microstructures developed in Zr-alloys during irradiation are complicated [5] therefore, it is important to review the impact of radiation damage on the safety of nuclear power plants. Extensive studies have been carried out to investigate and characterize the radiation induced changes in the microstructure of neutron irradiated Zr and Zr alloys [6] however, there are very little data on behavior of -Excel alloy in irradiation environment. For successful prediction and assessment of pressure tube performance in SCWR over a long duration in irradiation environment, it is crucial to understand the microstructural evolution mechanism in this material. A wide range of well controlled experimental conditions are required to study the basic damage process which is not readily achievable during neutron irradiation experiments [7]. This can be done by in-situ ion irradiation experiments in transmission electron microscope (TEM).

In the present study, we have investigated the evolution of microstructure in Excel alloy using Kr^{+2} ion irradiation at 100, 300 and 400°C. Damage microstructures have been characterized by TEM after a dose level of 10 dpa in both alpha and beta phases. Irradiation induced precipitation and growth of ω- phase has been observed. Furthermore, formation and growth of c-type loops, which are responsible for accelerated breakaway growth in Zircaloys [6] have been observed and recorded. This study provides an insight into the radiation induced microstructural evolution of Zr-Excel alloy, which can help to improve the design of Generation-IV SCWR-CANDU pressure tubes.

2. Experiments and Methods

The material used in this study is an Excel alloy pressure tube, which was provided by AECL Chalk River Nuclear Laboratories. As-received material has a dual phase microstructure comprising of α-hcp and β-bcc. The as-received material was heated to a temperature of 850oC in an argon environment for two hour and subsequently water quenched.

For microstructural analysis in TEM, specimen were punched from thin foils of heat treated material then electropolished in a solution of 5.3g LiCl, 11.16g Mg(ClO4)2, 100ml Butyl cellosolve, and 500 ml Methanol at -45°C in a TenuPol-5™ twin-jet electro-polisher. These specimens were analyzed in PHILIPS CM-20 at an operating voltage of 200 KV. Heat treated samples show 60% alpha and 40% beta phase. ω-phase was not observed in the bright field (BF) or dark field (DF) TEM micrographs; however, selected area diffraction (SAD) patterns show the presence of ω- phase. ω-phase present after the heat treatment is perhaps present in form of very fine particles with size less than 0.5 nm [8,9]. Fig. 1 shows the microstructure of the as received pressure tube material, heat treated material and SAD pattern from the β-phase of heat treated material.

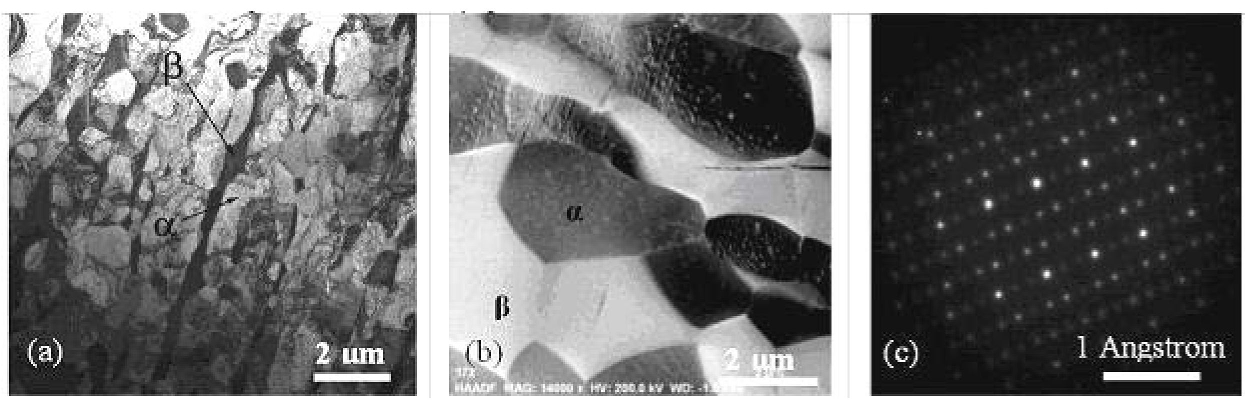

Fig. 1. (a) BF micrograph showing as received material microstructure, (b) HADF z-contrast image showing microstructure of heat treated material. (c). SAD pattern showing extra reflections from ω- phase.

Ion irradiation experiments were carried out at the IVEM-Tandem Facility at Argonne National Laboratory. The Facility includes a Hitachi H-9000NAR transmission electron microscope interfaced to a 2MV tandem ion accelerator. TEM samples were mounted in a double-tilt heating holder and irradiated at 300, 400, and 500^{o}C with 1 MeV Kr^{+2} ions to maximum total dose of 10 dpa. The dose rate was typically about 10^{-3} dpa/s according to SRIM 2008 calculations with displacement energy 40 eV. All in-situ dynamic TEM observations of damage evolution were performed at 300 KeV operating voltage which is well below the threshold voltage for electron knock-on damage in Zr. Damage microstructures were characterized under weak beam dark field (WBDF), kinematical bright-field (KBF) and dynamical two-beam diffraction conditions. In in-situ TEM analysis, evolution of microstructure was recorded in the same area of each specimen.

3. Results and Discussion

Some major observations are here briefly described at three irradiation temperatures, T_{irr}=400, 300 and 100^{o}C.

3.1. Irradiation Temperature (T_{irr})=400^{o}C

To investigate the nucleation and growth of c-type loops at 400^{o}C, an in situ irradiation experiment was carried out under the diffraction condition 0002[$11\bar{2}0$]. Pre-irradiation and final dose microstructures have been shown in fig. 2.

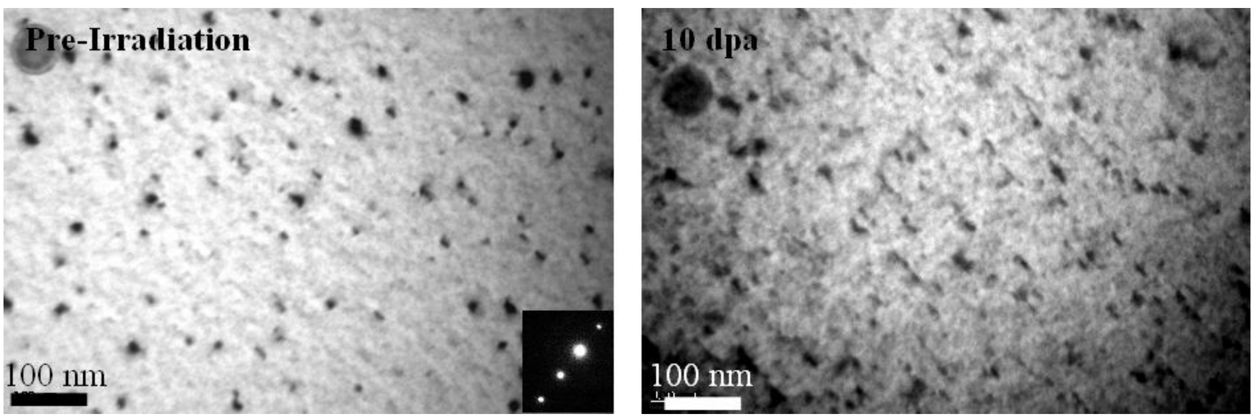

Fig. 2. Formation of c- loops in α-phase; c-loops nucleate and grow in the vicinity of SPP at 400^{o}C and 10 dpa. SPP disappear as the loops grow. g=0002, B~[$11\bar{2}0$].

The pre-irradiation microstructure shows the presence of small precipitates which are perhaps secondary phase particles (SPP). These SPP's are observed to dissolve due to the combined effects of irradiation and temperature. Dissolution of SPP decreases the stacking fault energy of the Zr which in turn enhances the nucleation of c-loops above a certain dose level [6]. After 2.5 dpa, c-type loops start to appear accompanied by the shape change of SPP's. c-loops are formed in the form of thin line segments in the vicinity of SPP's which grow with the increment of dose. Sudden increase in c-loops density above a threshold dose is considered to be the main cause of accelerated breakaway growth observed in Zircaloys [6]. c-loop formation in the form of short line segments is consistent with that observed after neutron irradiation. It has been reported that in neutron irradiation, this kind of loops has a Burgers vector of $\frac{1}{6}\langle 20\bar{2}3\rangle$ and are invariably vacancy in nature [6].

Formation of ω-phase has also been observed in metastable β-phase after the irradiation. ω-phase has been observed to precipitate preferentially at the interface of α and β phase. Fig. 3 shows the formation of cube shaped particles of ω- phase decorated at the boundary of β-phase present in α-matrix. Some of the ω-phase has also been observed at the core of beta phase.

It is notable that orientation relationship between ω/β is same as α/β [8], given by the burgers relationship $\{111\}_{\beta}\|(0001)_{\omega};\langle 1\bar{1}0\rangle_{\beta}\|\langle 11\bar{2}0\rangle_{\omega}$. ω-phase grows during irradiation maintaining this Burgers relationship, which implies that coherency of ω/β is not disturbed at the three irradiation temperatures.

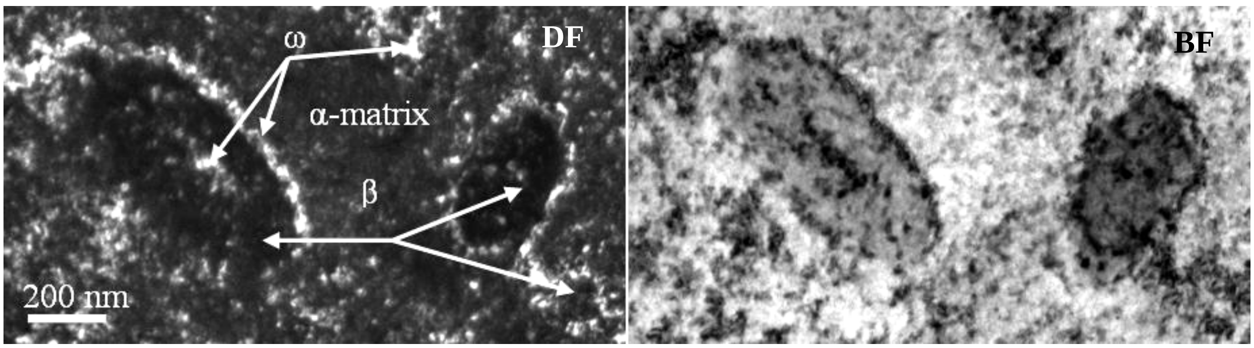

Fig. 3. showing the precipitation of growth of ω-phase in the small β-phase islands present in α-matrix. Cube shape ω-phase precipitates and grow preferentially at the α/β boundary. Some particles are present in the centre of β-phase islands.

3.2. T_{irr}=300^{o}C

Ion irradiation experiment at 300^{o}C shows similar features as observed at 400^{o}C as shown in fig. 4. Formation of c-loops has been observed ex-situ after a dose of 10 dpa fig. 4(a). Morphology of ω- phase at 300^{o}C resembles that observed at 400^{o}C i.e. cubic particles fig. 4(b).

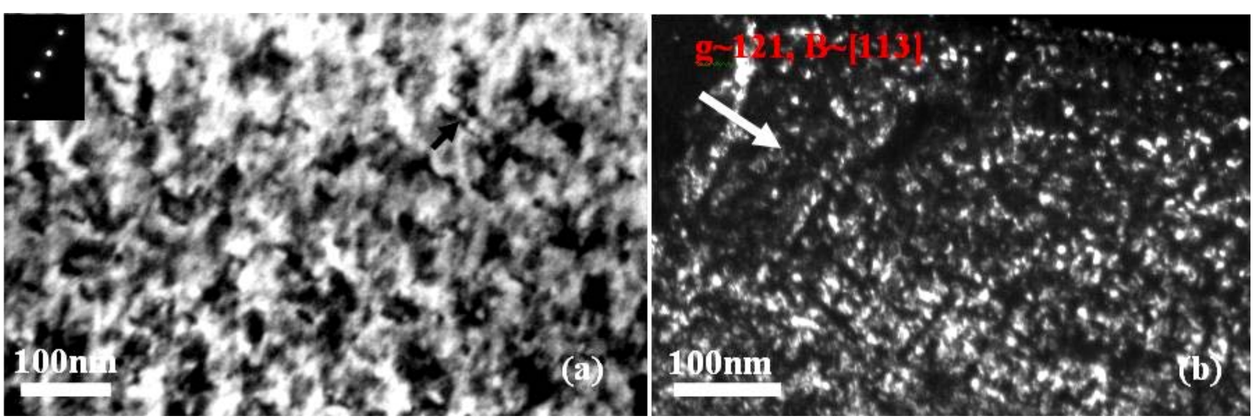

Fig. 4. (a). shows the presence of c-loops in form of thin short segments (arrowed) in α-phase at 300°C at a dose level of 10 dpa, g=0002 B~[11$\bar{2}$0] (b). Cube shape ω-phase precipitates similar to those observed at 400°C.

3.3. T_{irr}=100°C

At 100°C TEM micrographs show the presence of c-type loops after a dose of 10 dpa. c-type loops appear as short segments aligned parallel to the trace of basal plane, and are uniformly distributed throughout the grains as shown in fig. 5(a).

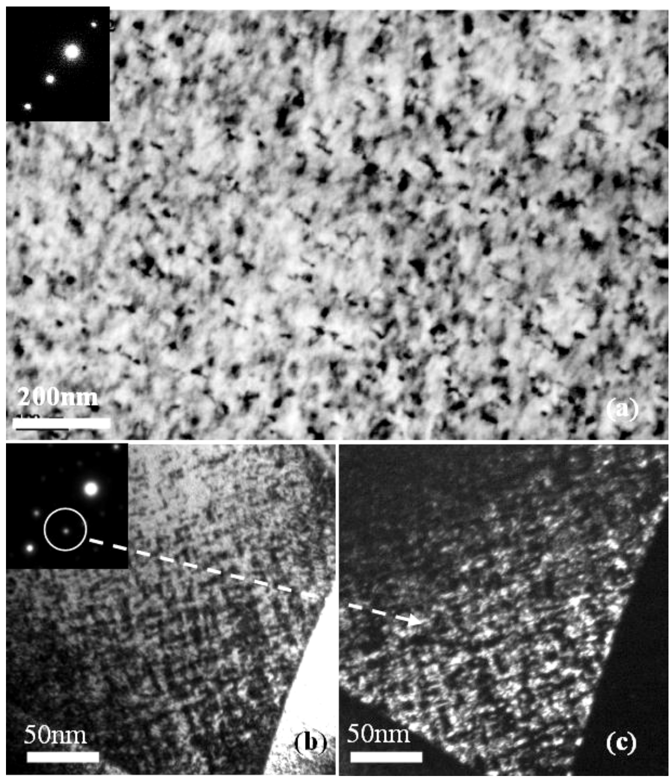

Fig.5. Major observations at 100°C after irradiations to 10 dpa. (a) shows the uniform distribution of c-loops in α-phase. Length of the segments is decreased as compared to 400°C.g=0002, B~ [11$\bar{2}$0] (b). BF micrograph showing the presence of ω-phase arranged in rectangular patterns, which is visible in form of thin Plates when imaged by selecting the ω-reflection (encircled) in SADP, g=$\bar{2}$11 B~[011] (c).

The most interesting observation during the irradiation experiment at 100°C is perhaps the appearance of ω-phase in the form of thin plates (fig.5c), in contrast to the cubic morphology observed at 400 and 300°C. These plates are arranged in the form of rectangular arrays.

4. Conclusion and Future Work

1- Heavy ion irradiation experiments on Zr–Excel alloy have revealed several new and interesting observations. Formation of basal plane c-loops above a threshold dose in the form of thin line segments is consistent with the previous observations in neutron irradiated Zircaloy -2.

2- c-loops are formed preferentially on the pre-existing SPP's present in the matrix. These SPP's change their shape and then dissolve as the c-type loops grow.

3- ω-phase formation has been observed at all temperatures; however morphology of ω- phase varies with temperature. At 400°C, ω-phase shows cubic morphology whereas at 100°C it appears as thin plates.

4- Effect of irradiation induced ω-phase formation on the mechanical properties of Zr-Excel alloy is not known yet. Further experiments are required to investigate the resultant mechanical properties of Zr-Excel alloy.

5. Acknowledgment

This work is sponsored by NSERC, UNENE (COG, OPG) and Nu-Tech Precision Metals under the Industrial Research Chair Program in Nuclear Materials at Queen's University. The ion irradiation was accomplished at the electron Microscopy Centre for Materials Research at Argonne National Laboratory, a US Department of Energy Office of Science Laboratory under Contract No. DE-AC02-06CH11357 by UChicago Argonne, LLC. The authors are grateful to Dr. Mark Kirk and Pete Boldo of Argonne National Lab for their help on the ion beam facility.

6. References

1- E.F. Ibrahim, E.G. Price and A.G. Wysiekiersky, 11(1972) 273.
2- B.A. Cheadle, R.A. Holt, V. Fidleris, A.R. Causey and V.F. Urbanic, Zirconium in the Nuclear Industry, Fifth International Symposium, ASTM, STP 754(1982)193-207.
3- A.R. Causey, G.J.C. Carpenter and S.R. MacEwen, Journal of Nuclear Materials 90(1980)216-223.
4- C.R.F. Azevedo, Engineering failure Analysis, 17(2011)1921-1942.
5- M. Griffiths, Philosophical Magazine A, 63(5) (1991) 835-847.
6- M. Griffiths, Journal of Nuclear Materials 159 (1988) 190-218.
7- G.S. Was, Fundamentals of Radiation Materials Science: Metals and Alloys, Springer, (2007)
8- S. Banerjee, P.Mukhopadhyay, "Phase Transformation: Titanium and Zirconium Alloys", Elsevier Science, 1 edition (2007).
9- S. Banerjee, R. Krishnan, Acta Metallurgica, 19(1971)1317-1326.