Study of irradiation induced defects and phase instability in β phase of Zr Excel alloy with in-situ heavy ion irradiation

Hongbing Yu[1], Zhongwen Yao[1], Mark A. Kirk[2], Mark R. Daymond[1]

(1) Department of Mechanical and Materials Engineering, Queen's University, Kingston, ON, Canada, K7L 3N6 (12hy1@queensu.ca)

(2) Material Science division Argonne National Laboratory, Argonne, IL, 60439, USA

Abstract

In situ heavy ion irradiation with 1 MeV Kr^{2+} was carried out to study irradiation induced phase change and atomic lattice defects in theβ phase of Zr Excel alloy. No decomposition of β-Zr was observed under irradiation at either 200 °C or 450 °C. However, ω-Zr particles experienced shape change and shrinkage associated enrichment of Fe in the β/ω interface at 200 °C irradiation but not at 450 °C. The defectevolution in the β-phase was examined with single phase Zr-20Nballoy. It was found that dislocation loops with Burgers vector ½<111> and <001> both present in β-Zr under room temperature irradiation.

1. Introduction

Zr Excel alloy (Zr-3.5Sn-0.8Nb-0.8Mo, wt.%) which has been proposed as candidate material of pressure tubes for CANDU-SCWR is a dual-phase alloy containing primary hcp α-Zr and metastable bcc β-Zr. Hexagonal ω-Zr phase could form in β-Zr as a result of aging during the processing of the tube. Extensive research work has been done on the counterpart of Zr Excel alloy, Zr-2.5Nb, showing that the metastable β-Zr (Zr-20 wt. %Nb) experiences decomposition and elements redistribution under irradiation environment [1-7]. However, little research has been done for the irradiation properties of the metastable β phase in Zr-Excel so far, thus leaving several problems unaddressed. 1) The stability of the minor phases in Zr-Excel alloy under irradiation; 2) the elemental redistribution induced by irradiation in the β-Zr and the ω-Zr phases; 3) irradiation induced defects in bcc β-Zr phase. In this study, in-situ heavy ion irradiation with 1 MeV Kr^{2+} at two different temperatures was carried out to investigate the stability of β-Zr and ω-Zr phases under irradiation. Qualitative Chemi-STEM was used to map alloying element distribution before and after irradiation. Due to the presence of ω particles which cause difficulties in characterizing small defects induced by irradiation, single phase bcc Zr-20Nb, which has similar lattice parameter with theβ-Zrin Zr Excel [8], was used as a model alloy to study the accumulation behavior of irradiation induced dislocation loops in β phase of Zr Excel alloy.

2. Materials and Experiment

Material used in this study is an as-received Zr-Excel alloy pressure tube material provided by Atomic Energy Canada Limited (AECL) Chalk River Laboratory with specified chemical composition give in table 1. The as received tube was fabricated by hot extruding the hollow billet at 850°C at a ratio of 10:1 followed by 25% cold drawing. The tube was then annealed at

750°C for 30 min and stress relieved at 400°C for 24 h. Zr-20Nb material used in this studymaterial is a 35% cold swaged rod which is 5mm in diameter. It was heat treated at 875 °C for 20 min to reduce the dislocation density and quenched in water to avoid the formation of ω phase and retain the bcc β phase structure.

Table 1The chemical composition of Zr Excel alloy in wt.%.

	Sn	Mo	Nb	O	Fe	H	Zr
Nominal	3.5	0.8	0.8	0.13	0.15	20ppm	Balance
Measured	3.39-3.68	0.77-0.81	0.75-0.77	0.11	0.09-0.13	5ppm	Balance

In-situ heavy ion irradiation was carried out at the Intermediate Voltage Electron Microscope Tandem Facility (IVEM-Tandem) at Argonne National Laboratory. The facility includes a Hitachi H-9000NAR TEM interfaced with a 2 MV tandem ion accelerator. TEM specimen made by electropolishing was mounted on double tilt TEM holder and irradiated under 1 MeV Kr^{2+} at a flux of $0.6 \times 10^{12} cm^{-2} s^{-1}$ at 200°C and 450°C to 10 dpa for ZrExcel sample and at room temperature to 1.5 dpa for bcc Zr-20Nb specimen. Dynamical observation was made at an operation voltage of 300KV. Dark field formed by ω reflection was used to view the change of microstructure during and after irradiation. Chemi-STEM EDX (Energy dispersion X-ray) element distribution mapping and ex-situ TEM characterization were performed on FEI TECHNAI OSIRIS microscope.

3. Experiment results

Figure 1 shows a series of micrographs captured during in situ observation demonstrating the change of morphology of the ω particles under irradiation up to 3 dpa at 200 °C. It can be clearly seen from figure 1 that ω articles gradually experienced shrinkage under heavy ion irradiation at 200°C, especially after 1 dpa. Figure 2 shows the morphology of of ω phase particles before and after irradiation to 10 dpa at 200 °C and 450°C. It clearly illustrates that the shape of ω particles changed remarkably from cuboid to ovoid after irradiation at 200°C to 10 dpa. However, no significant change is observed in the morphology of ω particles when irradiated at 450 °C. The shape factor, areal size distribution and the mean size of ω particles before and after irradiation are shown in Figure 3. Shape factor, which is defined by the ratio of long axis to short axis of ω particles, is changed to 1.926 from 1.277 after irradiation at 200°C indicating ω particles become more elongated rather than cuboidal. The mean areal size of ω particles decreases by 42%, from 160.7 nm^2 to 91.9 nm^2, after 10 dpa irradiation to at 200°C. The shrinkage of the ω particles under irradiation at 200°C is further demonstrated by left shift of the frequency peak. Selected area diffraction pattern shows that no amorphizationoccurred after irradiation at 200°C, thus indicating that ω particles experienced dissolution during irradiation at 200°C. No significant trends of growth and shrink of ω particles are observed after irradiated at 450 °C because neither the mean size nor size distribution of ω particles is changed significantly. No decomposition of β-Zr phase was observed under irradiation at both 200 °C and 450 °C.

Figure 4shows the Chemi-STEM mapping of Fe in βphase before and after irradiation. A striking feature is observed in the sample irradiated at 200℃ to 10 dpa, that increased level of Fe is observed around the boundary of the β matrix and ω particles. The Fe enriched region forms a loop and hasa similar shape to the outline of ω particles in unirradiated sample. Nevertheless, it is not observed in sample irradiated at 450℃ where dissolution ofω particles does not takes place. It might indicated the dissolution of ω particles in the sample irradiated at 200℃ is associated with the redistribution of Fe induced by irradiation.

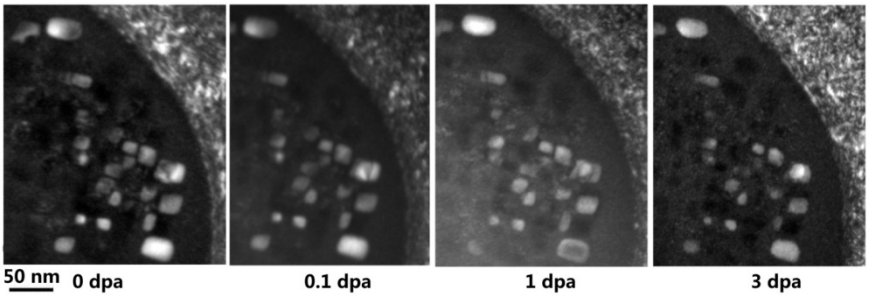

Figure 1In-situ observation of the changes of the morphology of ω phase particles under 1 MeV heavy ion (Kr^{2+}) at 200 ℃ up to 3 dpa. The dynamical observation is performed under TEM dark field formed withω diffraction spot.

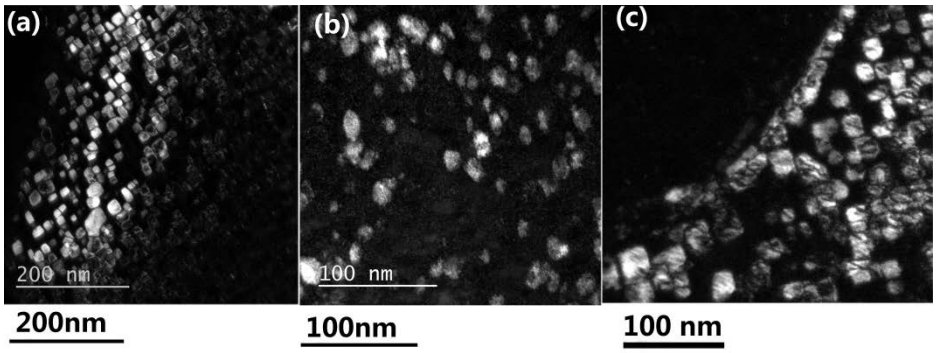

Figure 2The morphology of ω phase particles in samples, (a) unirradiated, (b) irradiated at 200℃ to 10 dpa, (c) irradiated at 450℃ to 10 dpa.

Figure 5 (a) and (b) show the typical dislocation loops induced by ion irradiation in β-Zr at room temperature to 1.5 dpa under and out of the illumination of electron beam, respectively. The mean size of the defects is about 6.2 nm under the illumination of electron beam and 4.7nm out of electron beam at 1.5 dpa. This indicates that the electron beam with which in-situ observation is carried out exhibits some influence on the growth of the dislocation loops. Figure 5(c) plots the number density of visible defects as a function of heavy ion dose in log-log scale. It illustrate that there is no visible defect yielded until 0.75 dpa, which is much higher than the threshold dose for visible defects in other bcc materials [9, 10] and hcp Zr [11]. The number density of dislocations could be expressed by an exponential function: $\rho = C(\phi)^n$, where ϕ is the ion dose, C is constant and n the exponential index, which is 3.52 and indicates that superimposition of 3

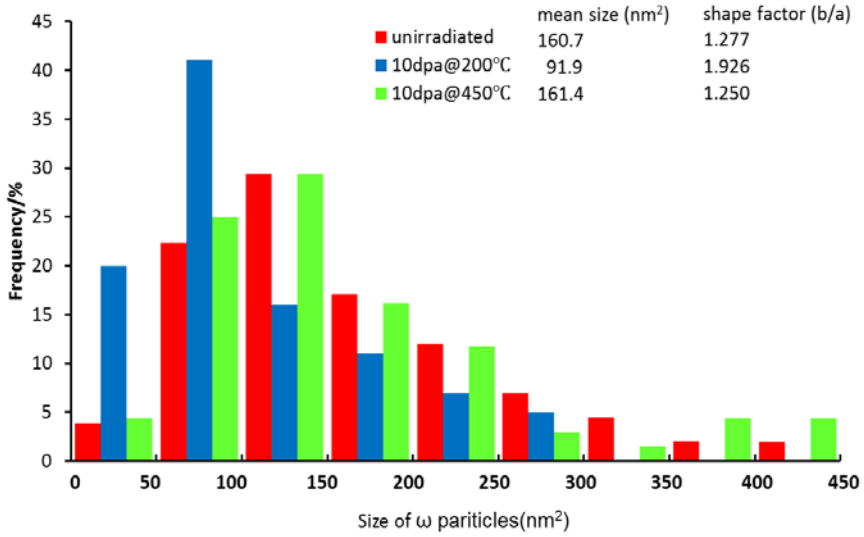

Figure 3Areal size distribution of ω particles before and after irradiation

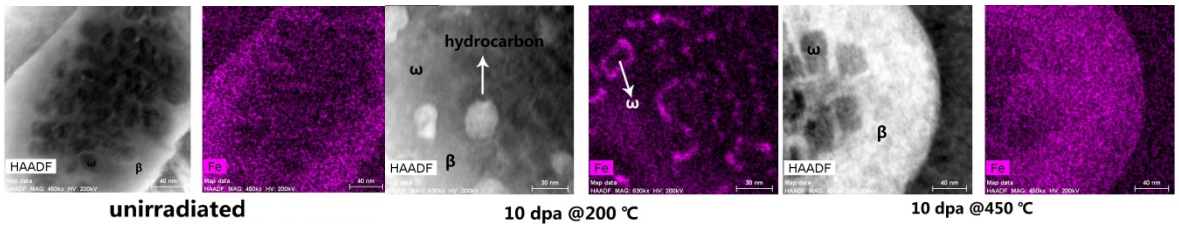

Figure 4The distribution of Fe in β phase before and after irradiation

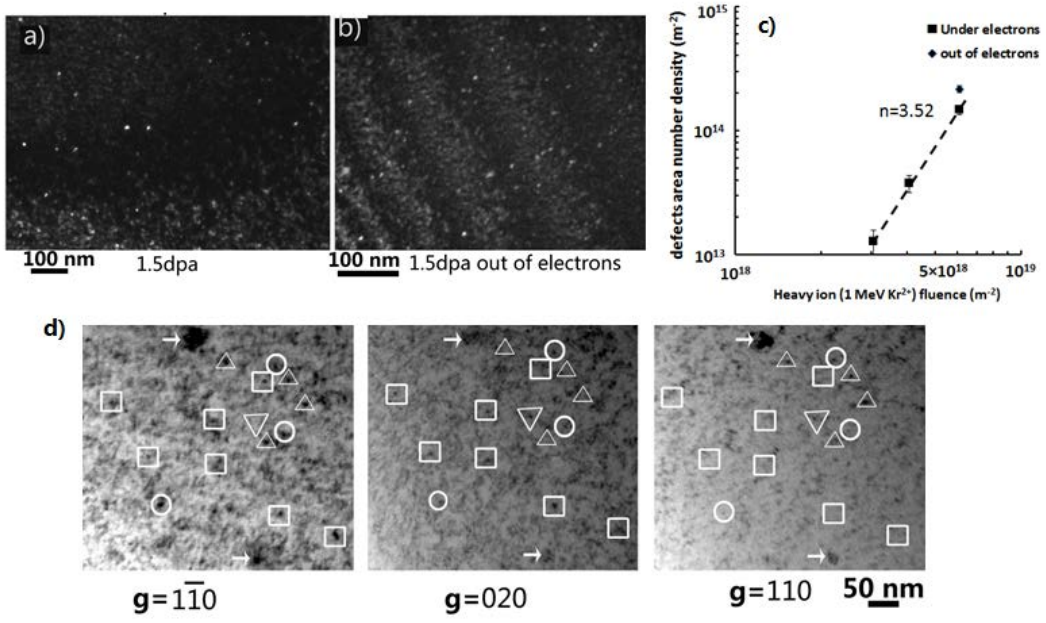

Figure 5 Defects accumulation behaviour in β-Zr at room temperature: (a) and (b) TEM dark field image of irradiation induced dislocation loops at 1.5dpa in the electron beam and out of electron beam, (c) loop number density as a f function of irradiation dose, (d) **g • b** analysis of dislocation loops.

or 4 cascades is required to produce one visible defect. Figure 5(c) shows a series of TEM bright field micrographs demonstrating the $\boldsymbol{g} \cdot \boldsymbol{b}$ analysis of the dislocation loops. According to the $\boldsymbol{g} \cdot \boldsymbol{b}$rule, loops which are visible in all three graphs have Burgers Vector [010] (loops which are circled). Loops which are marked with squares and only visible in $\boldsymbol{g} = 1\bar{1}0and\boldsymbol{g} = 020$have Burgers vector ½[111] or ½[11$\bar{1}$];those only visible in $\boldsymbol{g} = 1\bar{1}0$ and $\boldsymbol{g} = 110$and surround with triangles have Burgers vector [100]; the one only invisible in g =$1\bar{1}0$ which is surround by inverted triangle has Burgers vector ½[$\bar{1}$11]. This analysis means that dislocation loops with both Burgers vector ½<$\bar{1}$11> and <100>are induced by heavy ion irradiation in bcc β-Zr.

4. Conclusion

In situ heavy ion irradiation with 1 MeV Kr^{2+} was carried out in the β phase of Zr-Excel alloy and bcc Zr-20Nb alloy to investigate the stability of metastable minor phases in Zr Excel alloy and irradiation induced defects in bcc β-Zr phase, respectively. It was found that stability of ω particles depends on irradiation temperature. The ω-Zr particles experienced shape changes and eventual dissolution from outer layer to core associated with enrichment of Fe at the β/ω interface at 200 °C irradiation but not at 450 °C. No decomposition of β-Zr phase was observed under irradiation at either 200 °C or 450 °C. Dislocation loops with Burgers vector ½<111> and <001> were both observed in β-Zr under room temperature irradiation. The mean size of the defects is bout 5-6 nm, and the threshold dose for visible defect is much higher than that for its hcp counterpart.

5. Reference

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