CHARGED PARTICLE IRRADIATION EFFECTS ON ZIRCALOY-4

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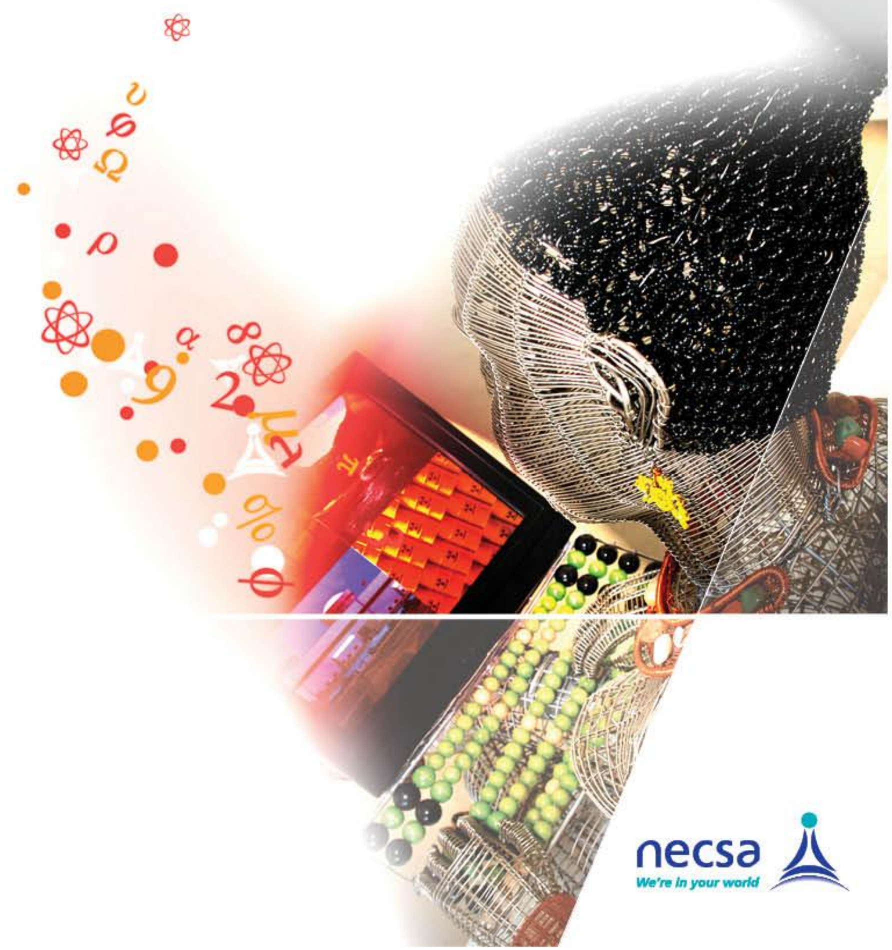

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INTRODUCTION

- During the service lifetime of reactors, key reactor materials such as zircaloy-4 degrade.

- Their degradation is due to the fast neutron environment generated in the reactor core during fission.

- This alters the physical and mechanical properties of zircaloy-4 resulting in the material becoming the limiting factor in continual operation of the reactor.

- To overcome this limitation, better radiation resistant materials need to be developed.

- Their development however requires a fundamental understanding of the mechanisms governing this degradation.

BURNUP INCREASE PER GENERATION

GWd/tU – GigaWatt day per ton of Uranium

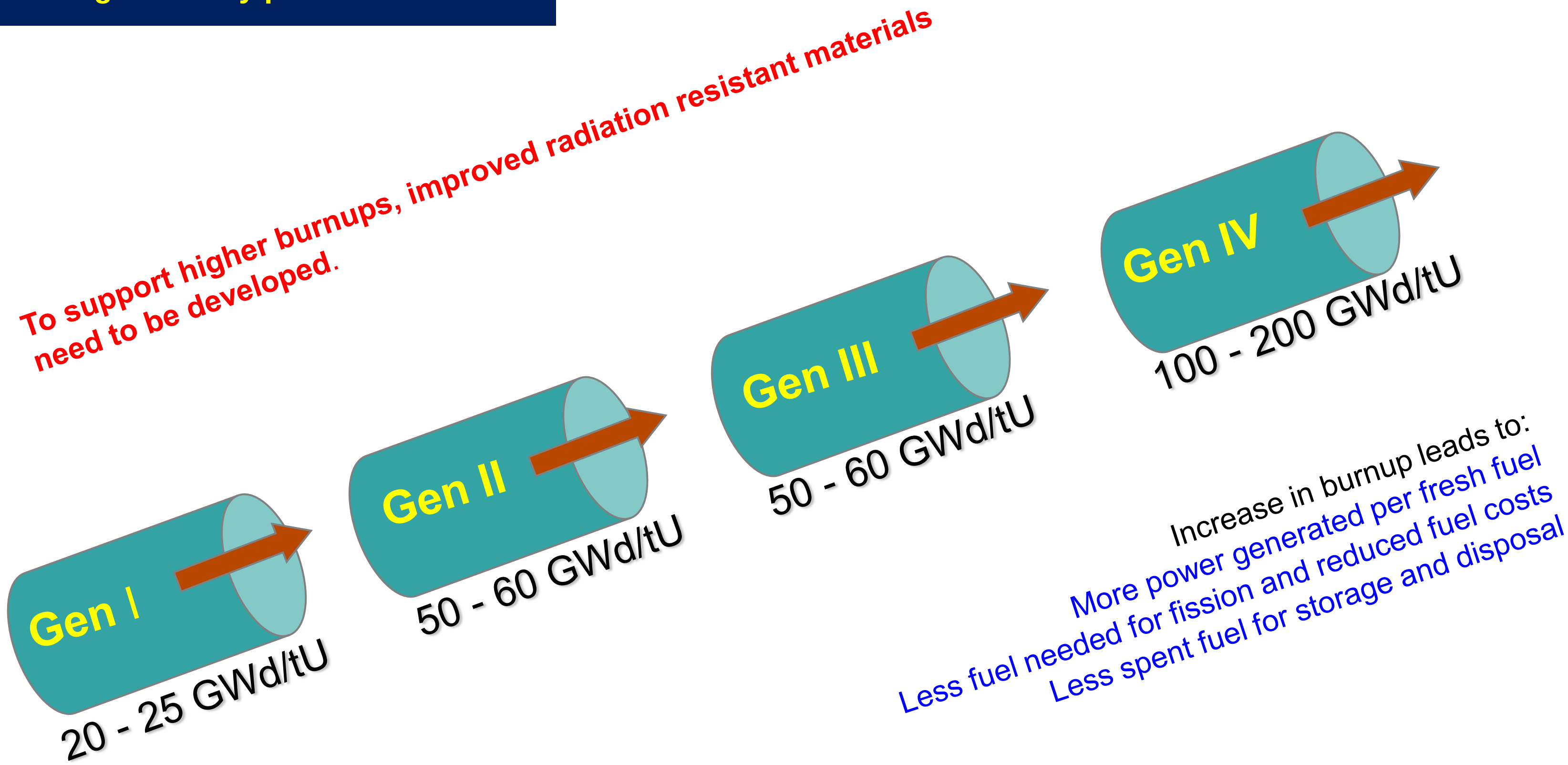

Figure 1: Schematic representation of burnup increase per generation of reactors

ZIRCALOY-4

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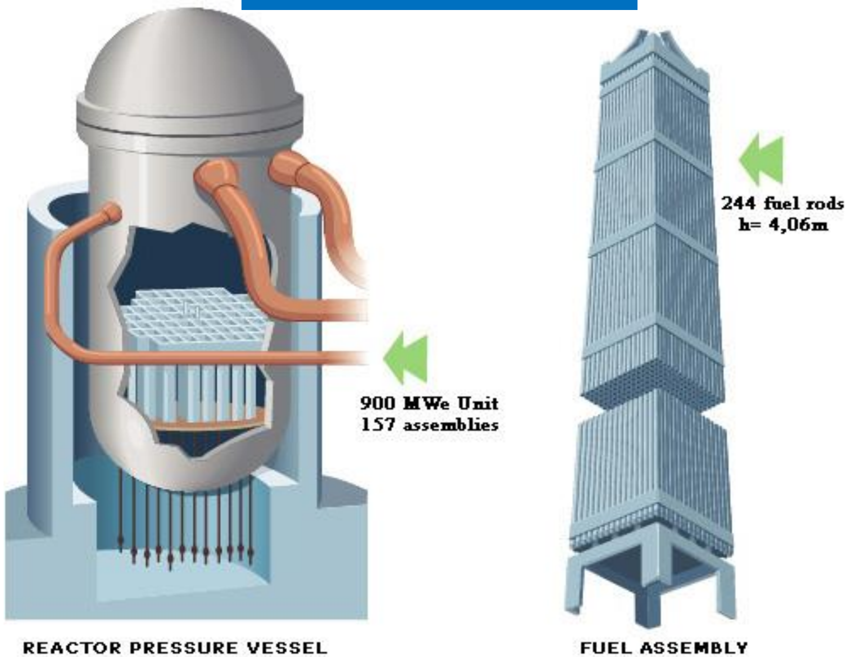


Figure 2: Schematic representation of a nuclear reactor core with fuel assemblies

- Tin (Sn) - 1.45 % wt
- Iron (Fe) – 0.2 wt %
- Chromium (Cr) – 0.1 wt %
- Zirconium (Zr) - balance

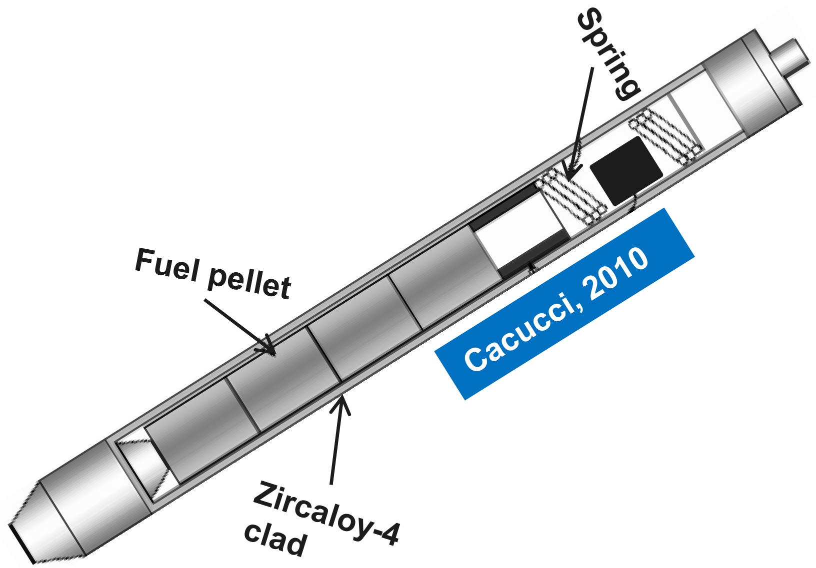


Figure 3: Cross section of a single fuel rod showing clad material

- Low neutron absorption cross section
- High mechanical strength
- High corrosion resistance
- High thermal conductivity

RADIATION DAMAGE PROCESS

B. Linn, 2013

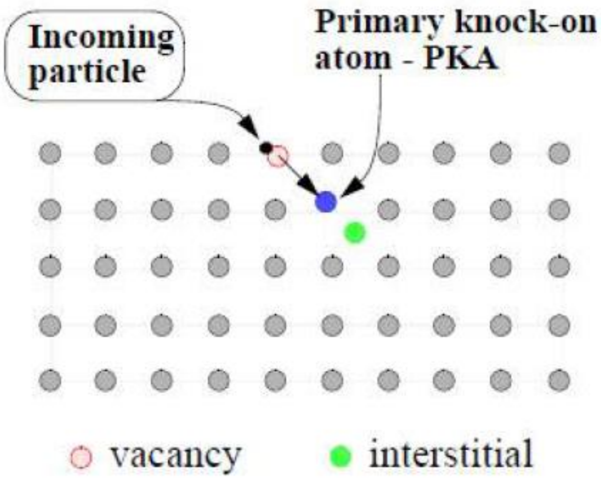


Figure 4: Schematic representation of radiation damage process

Table 1: Displacement threshold energy of elements

Element	E_d (eV)
C	30
Al	27
Si	25
Zr	40
Fe	40
Cu	40

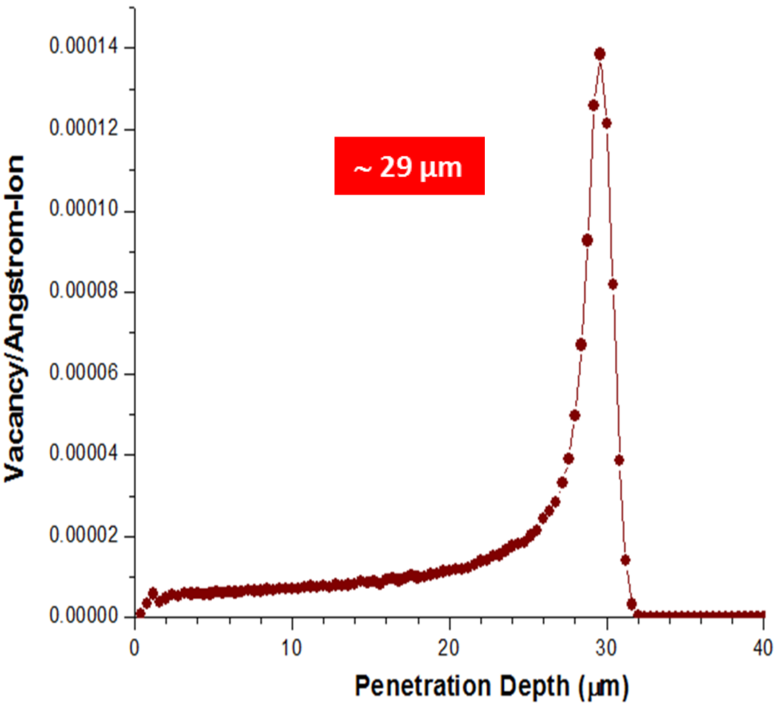


Figure 5: SRIM Simulation of the penetration depth of protons in zircaloy-4

SAMPLE PREPARATION AND CHARACTERIZATION

Polishing and Etching

- Sample polished used Silicon carbide (SiC) papers of various grit sizes (1200,2400, and 4000) and diamond paste of various roughness (1 µm and 0.25 µm).
- Sample etched with a mixture of 10% Hydrofluoric acid (HF), 45% Nitric Acid (HNO_3), and 45% Hydrogen Peroxide (H_2O_2) after polishing.

Characterization Techniques

- Optical Microscopy (OM)
- Scanning Electron Microscopy (SEM)
- X-ray Diffraction (XRD)

IRRADIATION CONDITIONS

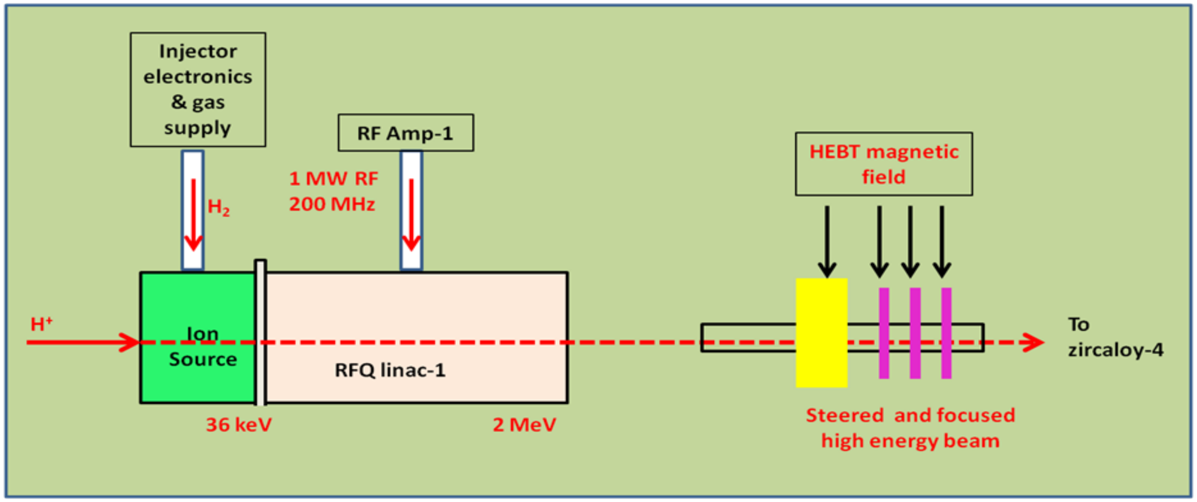


Figure 6: Schematic representation of a radio frequency quadrupole (RFQ) accelerator at Necsa.

Water cooled
Under vacuum at 10^{-7} mbar

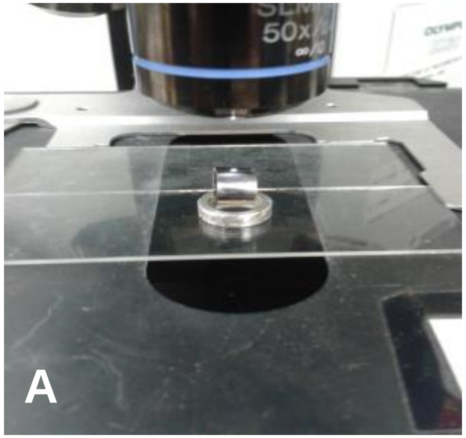
Table 2: Accelerator operation conditions during irradiation

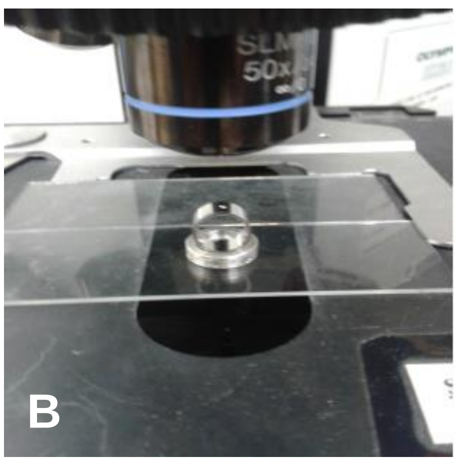
Parameters	Value
Energy (MeV)	2
Current (mA)	1.8
Beam spot (mm)	3
Pulse width (μs)	400
Repetition rate (Hz)	20

FLUENCE

1.2×10^{19} protons/cm^2

RESULTS: OM ANALYSIS





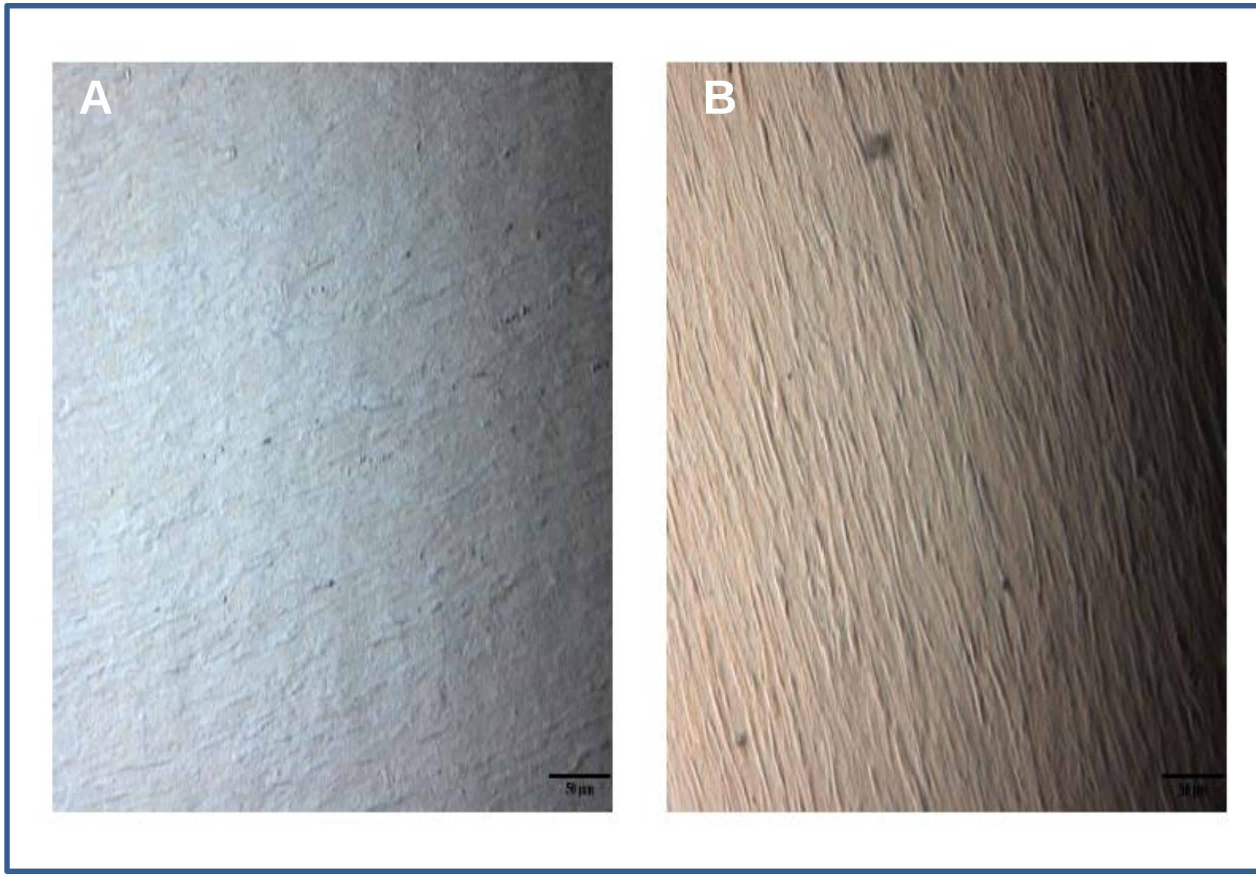


Figure 7: OM micrographs of virgin zircaloy-4 placed in the (A) horizontal direction and (B) in the vertical direction on microscope stage.

RESULTS: SEM ANALYSIS

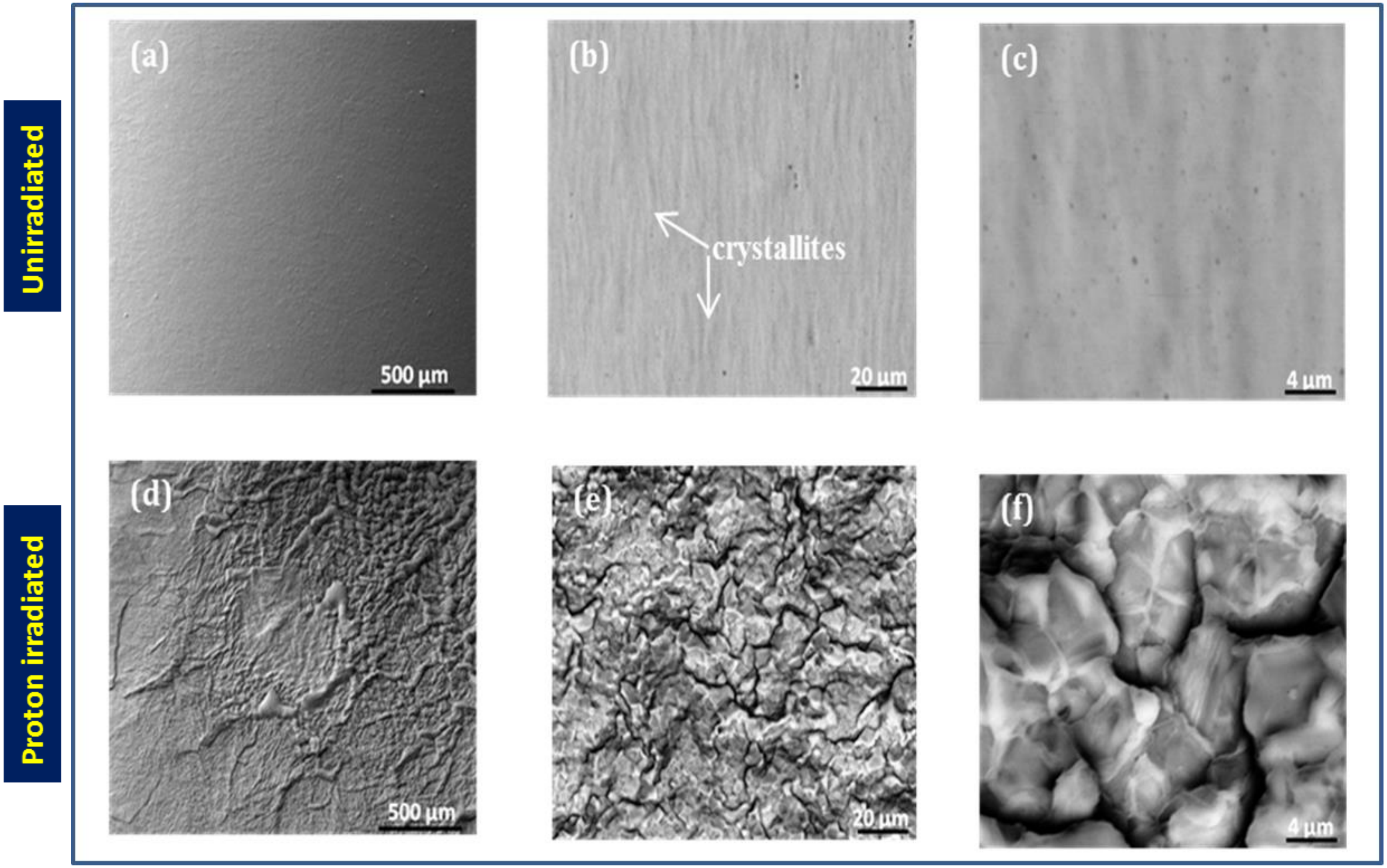


Figure 8: SEM micrographs of virgin ziircaloy-4 at (a) 120 X mag, (b) 2000 X mag, and (c) 10000 X mag and SEM micrographs of proton irradiated zircaloy-4 at at (d) 120 X mag, (e) 2000 X mag, and (f) 10000 X mag

RESULTS: XRD ANALYSIS

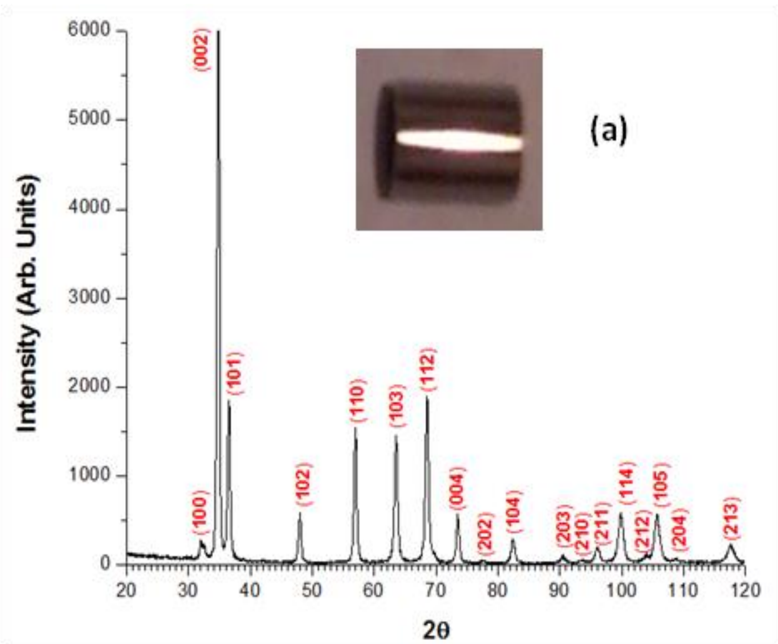


Figure 9: XRD pattern of un-irradiated zircaloy-4

Red Indices - Zirc-4
Blue Indices - ZrC

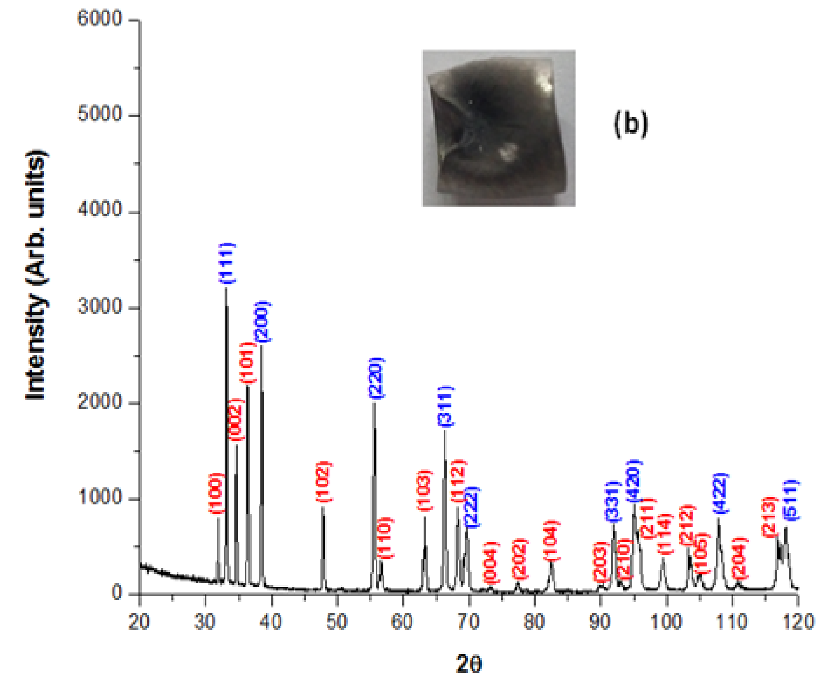
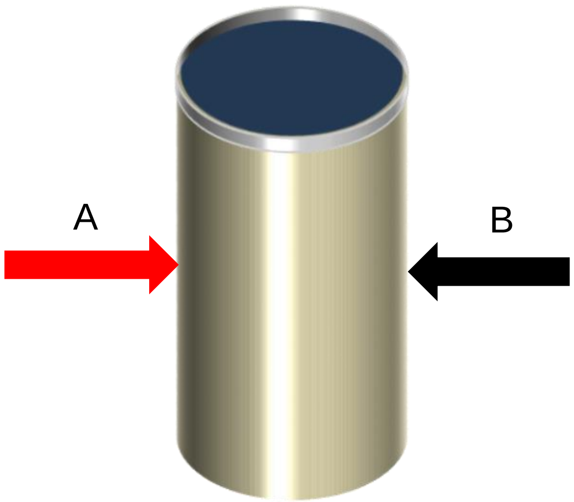


Figure 9: XRD pattern of proton irradiated zircaloy-4

RESULTS: RUTHERFORD BACKSCATTERING SPECTROSCOPY (RBS) ANALYSIS



A - Exposed
B - Unexposed

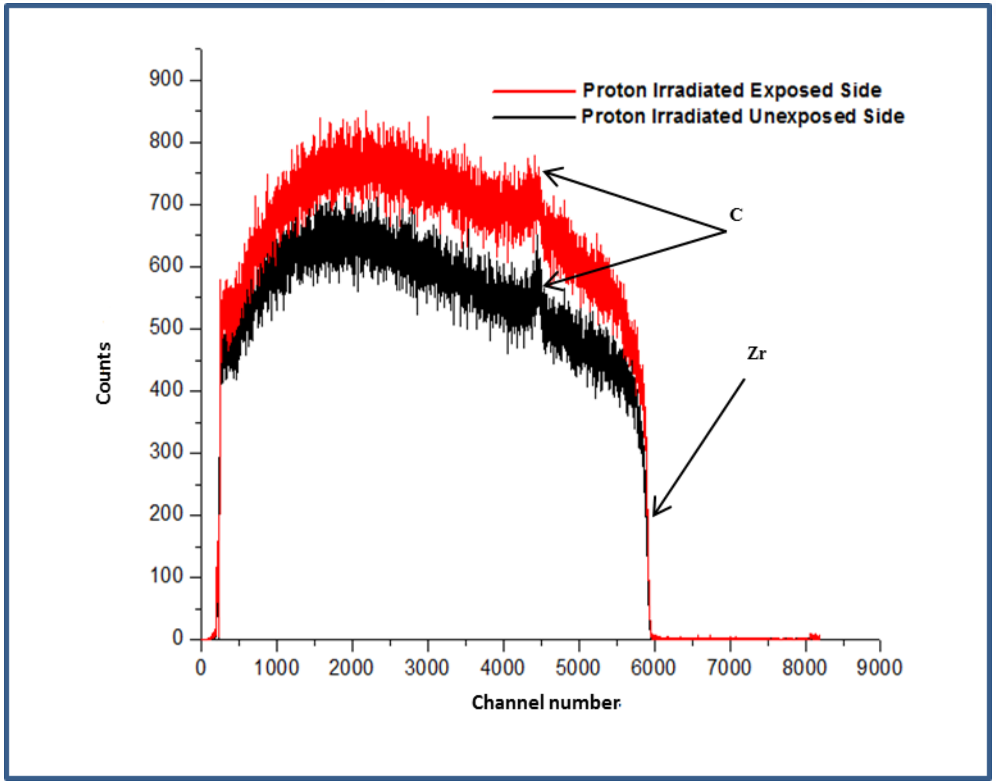


Figure 10: RBS spectra of unexposed (black) and exposed (red) proton irradiated zircaloy-4.

$$E_1 = kE_0 \qquad (1)$$

$$k = \left[\frac{(m_2^2 - m_1^2\sin^2\theta)^{\frac{1}{2}} + m_1\cos\theta}{m_1 + m_2}\right]^2 \qquad (2)$$

E_0 – incident particle energy

E_1 – backscattered particle energy

θ – scattering angle

m_1 – proton mass

m_2 – target mass

RESULTS: XRD PEAK COMPARISON

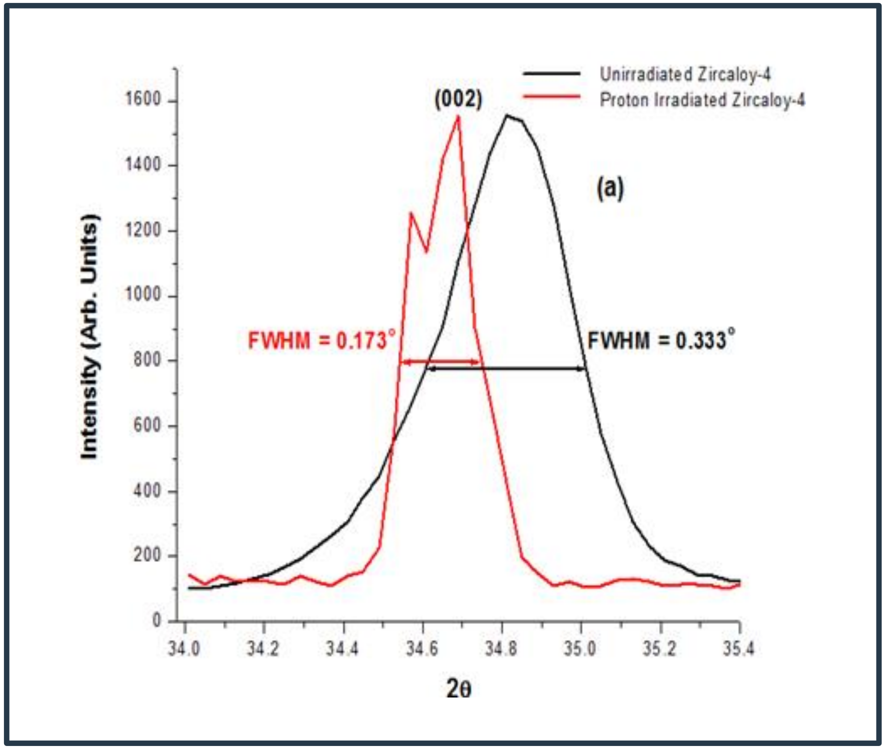


Figure 11: Comparison of the (002) peak of the unirradiated (black) and the proton irradiated (red) zircaloy-4.

$$d = \frac{\lambda}{2\sin\theta}$$

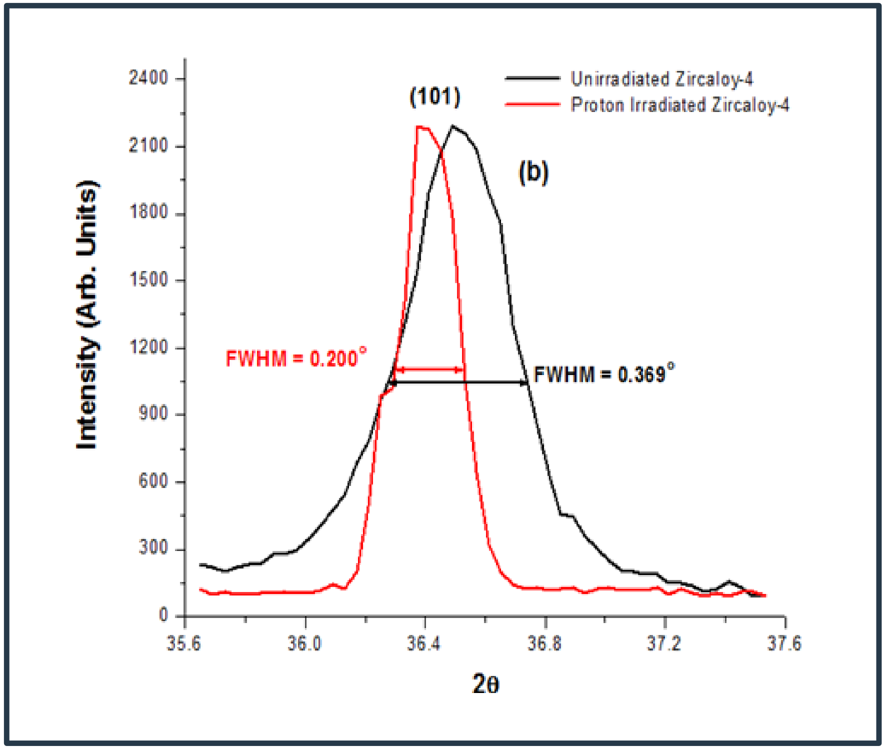


Figure 11: Comparison of the (101) peak of the unirradiated (black) and the proton irradiated (red) zircaloy-4.

$$L = \frac{K\lambda}{\beta\cos\theta}$$

RESULTS: XRD 2D COMPARISON

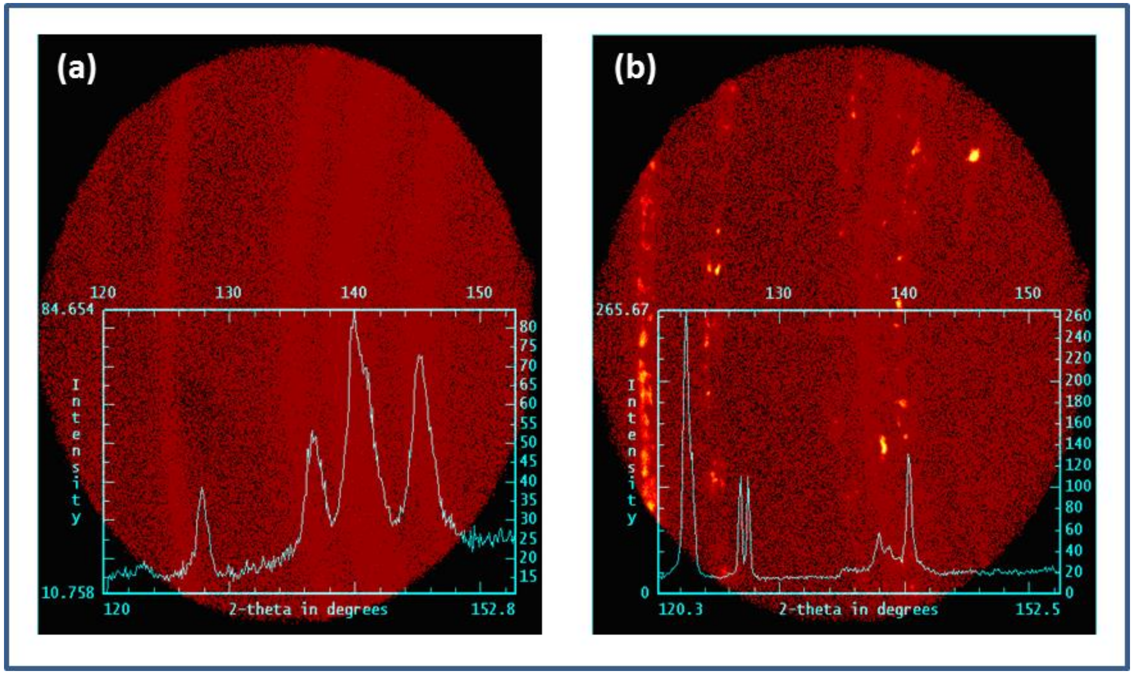


Figure 12: 2D XRD data frame of (a) un-irradiated and (b) proton irradiated zircaloy-4

Table 3: XRD quantitative analysis of zircaloy-4

Zircaloy-4 State	Residual Stress (MPa)		Interlayer spacing (A˚)		Particle Size (nm)
	σ_{11}	σ_{22}	$d_{(002)}$	$d_{(101)}$	
Virgin	-7.9 ± 1.0	-9.30 ± 0.9	2.577	2.460	35
Proton Irradiated	6.7 ± 2.2	8.0 ± 2.2	2.583	2.463	107

CONCLUSION

- Proton irradiation leads to changes in the microstructure of zircaloy-4.
- XRD and SEM revealed a change in crystallite size after proton irradiation.
- Melting and cracking on the surface of zircaloy-4 was observed with SEM.
- Residual stress reversal was observed with XRD even though it was within the instrument uncertainty.

Thank you