Effects of 710 MeV Bi Ion Irradiation and Annealing on Se-Implanted SiC

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Introduction

The growing global demand for clean and sustainable energy has intensified interest in nuclear power as a low-carbon alternative to fossil fuels. Despite its advantages, nuclear energy faces challenges related to the long-term storage of nuclear waste and safe containment of radioactive fission products (FPs), which can pose serious environmental and health risks if released.

Advanced nuclear reactor designs, such as the High Temperature Gas-Cooled Reactor (HTGR), employ tri-structural isotropic (TRISO) fuel particles (see **Fig. 1**) to enhance the safety and performance of nuclear fuel [1]. In these particles, the silicon carbide (SiC) layer acts as the primary diffusion barrier, preventing the escape of most FPs due to its excellent chemical stability and mechanical strength [2]. However, certain fission products, including iodine (I), silver (Ag), cesium (Cs), strontium (Sr), xenon (Xe), and krypton (Kr), have been found to escape the coating layers under extreme reactor conditions, posing potential environmental hazards [3].

selenium (Se), particularly the long-lived isotope Se-79, is another important FP which presents a radiological hazard owing to its beta emission. Understanding its retention and migration behaviour in SiC under reactor-like conditions is therefore crucial for improving fuel safety [4].

During fission reactions, energetic fragments with varying energies with some initially in the order of hundreds of MeV - comparable to swift heavy ions (SHIs), are generated and can significantly alter the microstructure of SiC and influence the migration of the pre-implanted species (see schematic diagram in **Fig. 2** showing a typical fission reaction) [5]. This study investigates the effects of SHI irradiation (710 MeV Bi^{+51}) and high-temperature annealing on the microstructural evolution and migration behaviour of Se-implanted SiC, thereby contributing to the understanding of fission product transport mechanisms in advanced nuclear fuels.

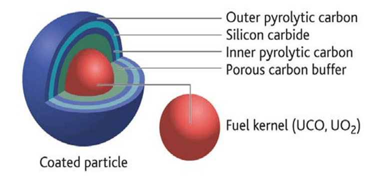

Fig. 1. Schematic representation of a TRISO particle.

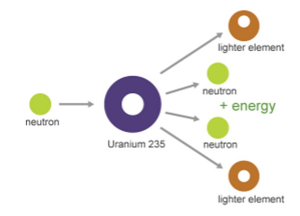

Fig. 2. Schematic representation of a nuclear reaction.

Experimental Procedure

Se ion implantation

- Substrates: Polycrystalline SiC
- Temperatures: RT and 350 °C
- Energy: 200 keV
- Fluence: 1×10^{16} cm^{-2}

710 MeV Bi^{+51} irradiation

- Substrates: Se pre-implanted SiC
- Temperatures: RT
- Energy: 710 MeV
- Fluence: 1×10^{13} cm^{-2}

Sequential annealing

- Temperatures: 1000 to 1300 °C
- Pressure: 10^{-6} Pa
- Time: 10 h

Analysis techniques

- Raman spectroscopy
- SEM
- RBS
- TEM

Simulations

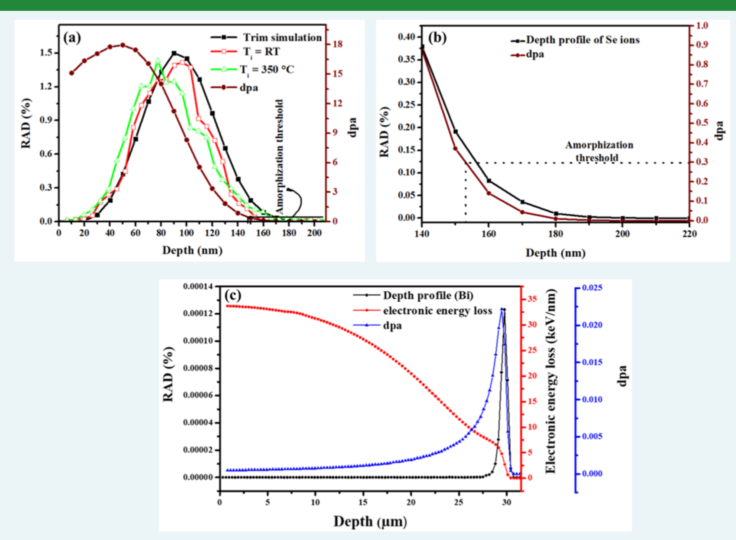

Fig.3. The relative atomic density (%) and displacement per atom (dpa) versus depth obtained from SRIM simulations and experimental results from RBS for (a) implanted Se ions to fluence of 1×10^{16} cm^{-2} at RT and 350 °C, (b) zoomed in version of (a) and (c) irradiated Bi ions to fluence of 1×10^{13} cm^{-2} and the electronic energy loss for 710 MeV Bi ions.

Results Cont...

RBS Results

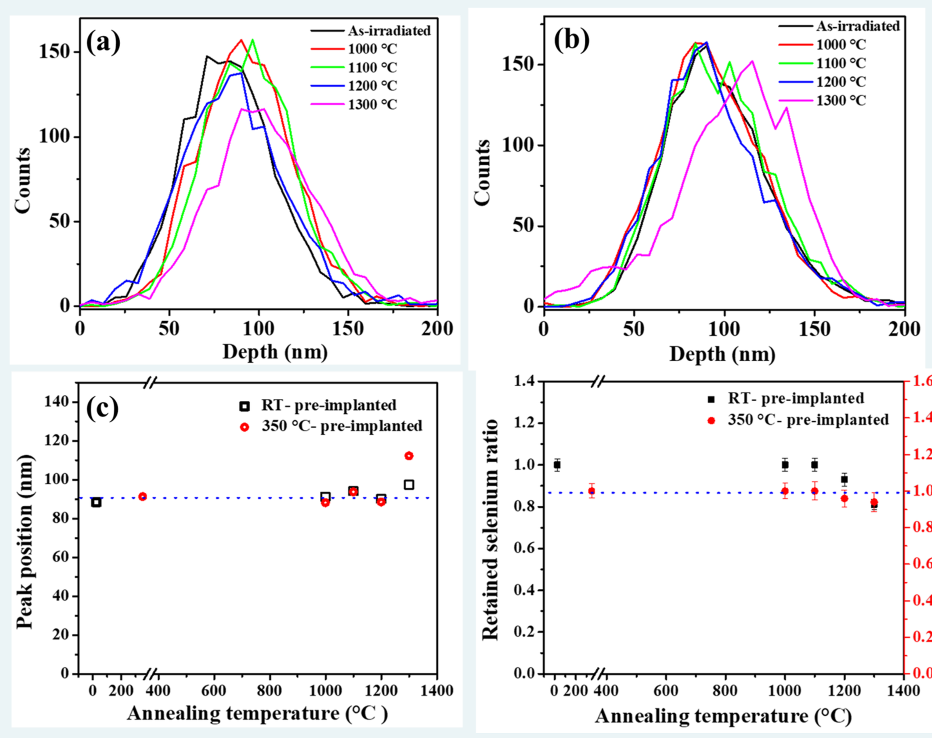

Fig.10. Se depth profiles from RBS of SiC pre-implanted at (a) RT and (b) 350 °C, then irradiated and annealed at 1000, 1100, 1200 and 1300 °C. (c) Se peak position and (d) retained Se as a function of annealing temperature.

Results

TEM Results (After Implantation & SHI Irradiation)

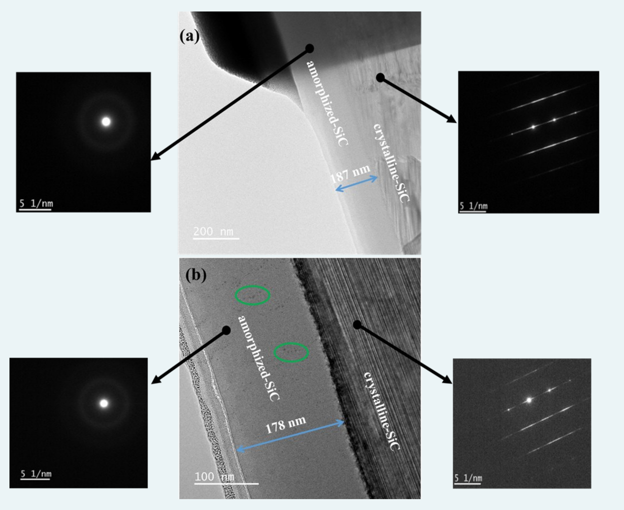

Fig.4. TEM micrographs of (a) as-implanted polycrystalline SiC at room temperature and subsequently (b) irradiated with SHIs. The SADs of amorphous and bulk (crystalline) are also incorporated.

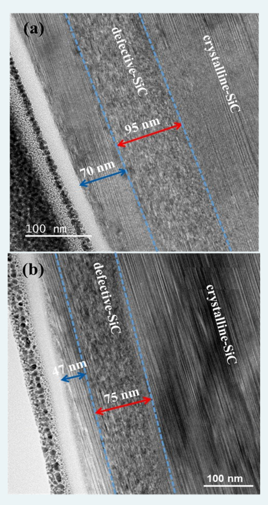

Fig.5. TEM micrographs of (a) as-implanted polycrystalline SiC at 350 °C and subsequently (b) irradiated with SHIs.

Raman Results (After Implantation & SHI Irradiation)

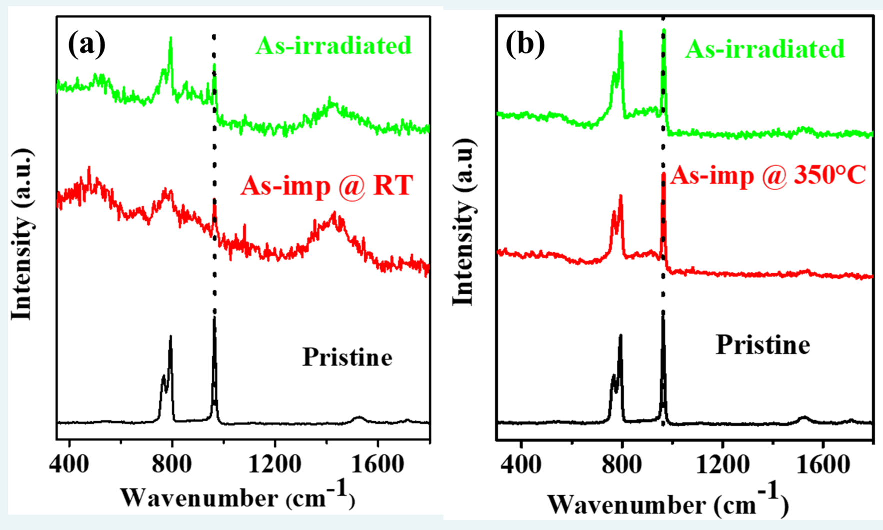

Fig.6. Raman spectra of the Se implanted samples at (a) RT, and (b) 350 °C and after irradiation. Pristine spectrum was included for comparison.

Raman Results (After Annealing (1000 - 1300 °C))

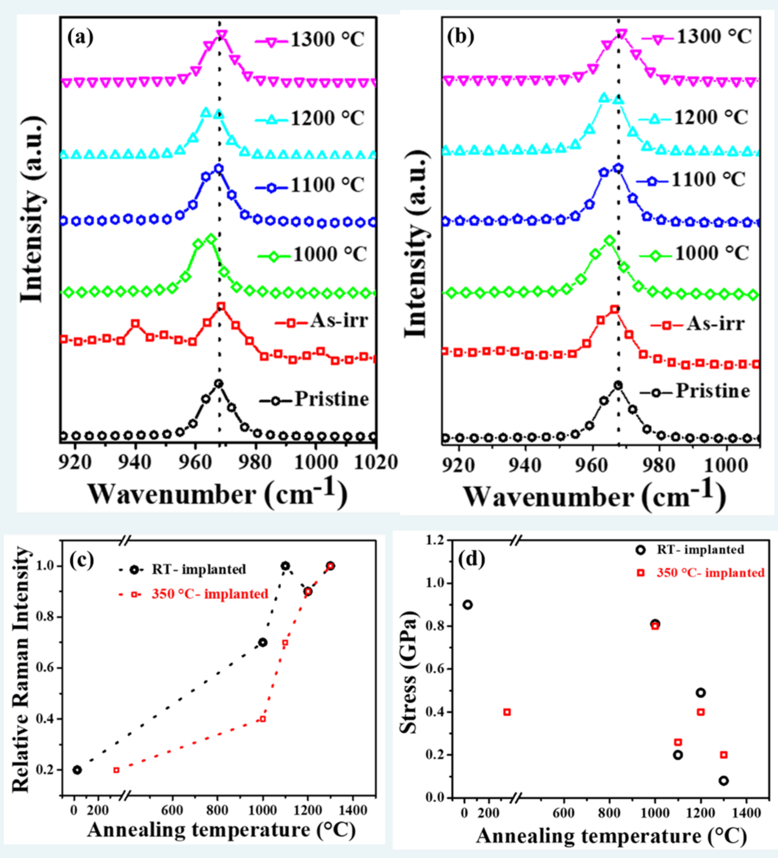

Fig.7. Magnified views of LO peaks extracted from Raman spectra of the pre-implanted SiC at (a) RT and (b) 350 °C then irradiated, subsequently annealed from 1000 to 1300 °C. (c) The relative Raman intensity (RRI) and (d) residual stress (GPa) versus the annealing temperature for SiC pre-implanted at RT and 350°C, followed by irradiation and annealing between 1000 and 1300°C. Pristine and as-irradiated samples are included for comparison.

SEM Results (After Annealing @ 1200 & 1300 °C)

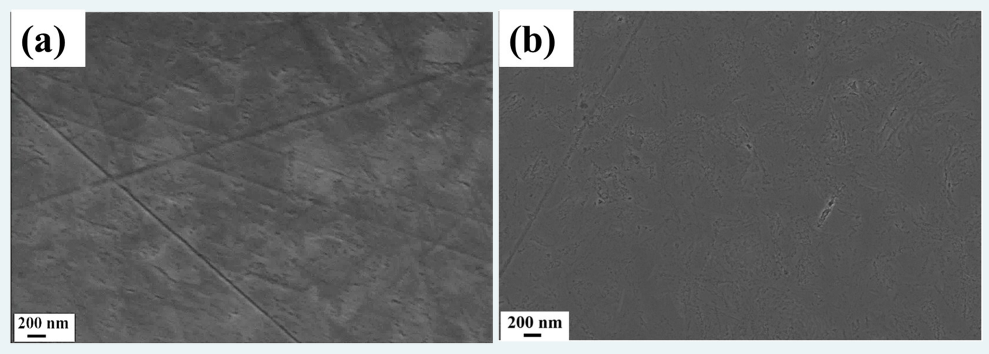

Fig.8. SEM micrographs of the RT pre-implanted SiC, then irradiated, subsequently sequentially annealed at (a) 1200 and (b) 1300 °C.

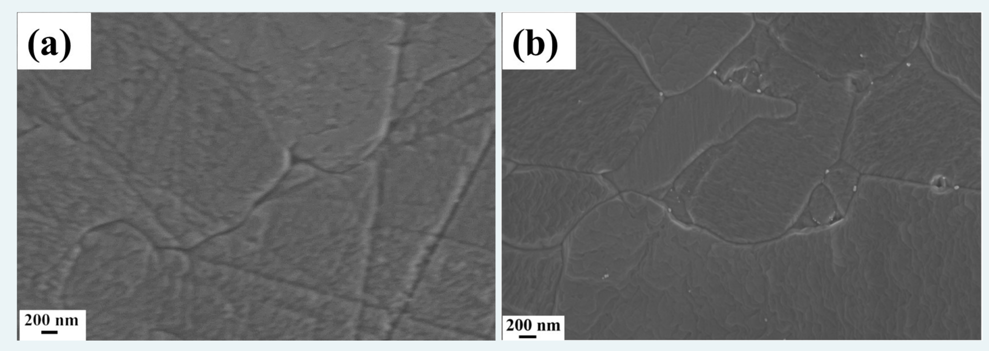

Fig.9. SEM micrographs of the 350 °C pre-implanted SiC, then irradiated, subsequently sequentially annealed at (a) 1200 and (b) 1300 °C.

TEM Results After Annealing @ 1200 °C

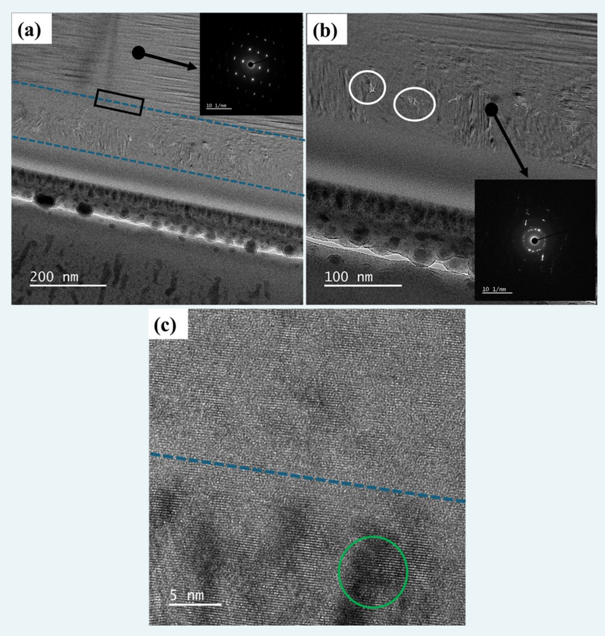

Fig.11. (a) BF-TEM, (b) higher magnification of (a) and, (c) HR-TEM micrographs of RT pre-implanted SiC then irradiated, subsequently annealed up to 1200 °C. The arrows point to the selected area diffraction (SAD) patterns captured from the defective and bulk-crystalline region.

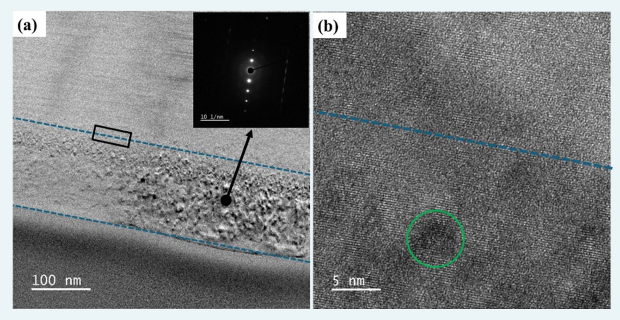

Fig.12. (a) BF-TEM and (b) HR-TEM micrographs of 350 °C pre-implanted SiC then irradiated, subsequently sequentially annealed up to 1200 °C. The arrow points to the selected area diffraction (SAD) patterns captured from the defective region.

Summary of Results

- RT Implantation of Se ions resulted in amorphization of SiC (retained amorphous layer of about 187 nm).
- Subjecting the amorphized layer to 710 MeV Bi^{51+} irradiation led to formation of randomly oriented nano-crystallites and reduction of the amorphous layer from 187 to 178 nm.
- Implantation at 350 °C retained SiC crystallinity, with a defective layer of about 165 nm, consisting of a less defective (~70 nm) and highly strained (~95 nm) layer.
- Irradiation of the defective layers induced some crystallization of defects (these layers shrank by 33% and 21%, respectively).
- Sequential annealing reduced defects in RT pre-implanted and subsequently irradiated sample, forming strained nanocrystalline SiC with cavities and Se precipitates at 1200 °C.
- While the samples pre-implanted at 350 °C produced nanocrystalline SiC with only minor strained regions.
- No detectable selenium migration occurred in either RT or 350 °C implanted samples after annealing up to 1200 °C.
- RT implanted samples showed ~20% Se loss, while 350 °C implanted sample showed significant Se migration into bulk SiC with minimal Se loss.
- Higher implantation temperature (350 °C) improved structural recovery and stabilized dopant (Se) distribution.

References

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Acknowledgements

► Financial support by the National Research Foundation of South Africa is gratefully acknowledged for funding this work
► iThemba LABS & JINR are acknowledged for sponsoring the attendance of this conference

NRF National Research Foundation

iThemba LABS Laboratory for Accelerator Based Sciences

JINR

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