संत लौंगोवाल अभियांत्रिकी एवं प्रौद्योगिकी संस्थान
(भारत सरकार द्वारा स्थापित)
Sant Longowal Institute of Engineering & Technology
DEEMED UNIVERSITY (Established by Govt. of India)

Study of zirconolite ceramic compositions through swift heavy ion irradiations from 15 UD tandem pelletron for immobilizing nuclear wastes

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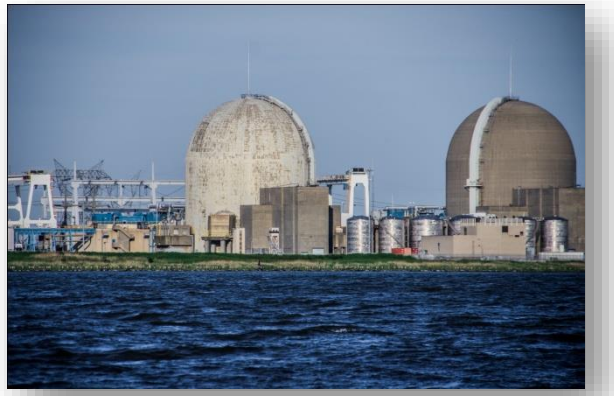
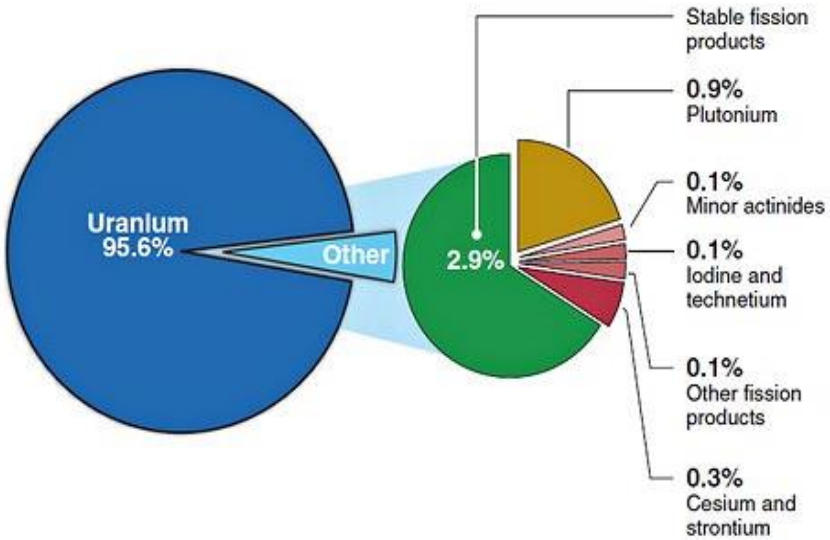
Outline

- Introduction
  - Nuclear Fuel & Waste
  - Indian Scenario
- Waste Management
- Waste Forms
- Zirconolite
- Radiation Effects in Materials
- Radiation Stability of Zirconolite
- Summary
- Acknowledgements

Nuclear Waste: A Dread



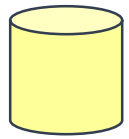
Nuclear reactor

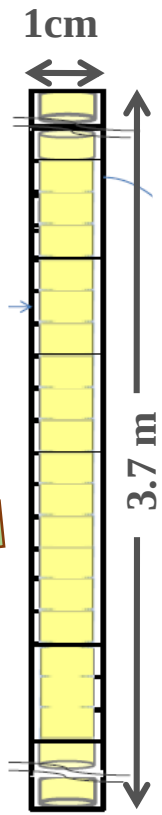


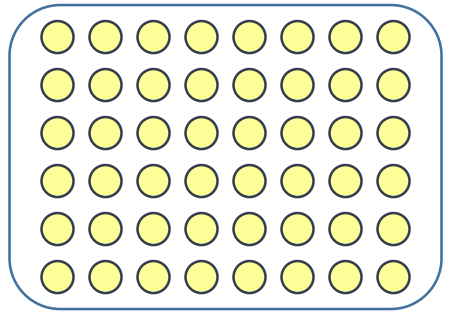
Spent Nuclear Fuel

Fuel assembly

UO_2
Fuel Pellet
(0.7% U^{235} & rest U^{238})



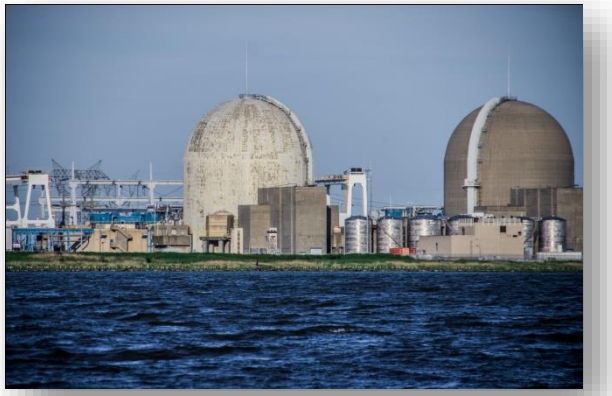




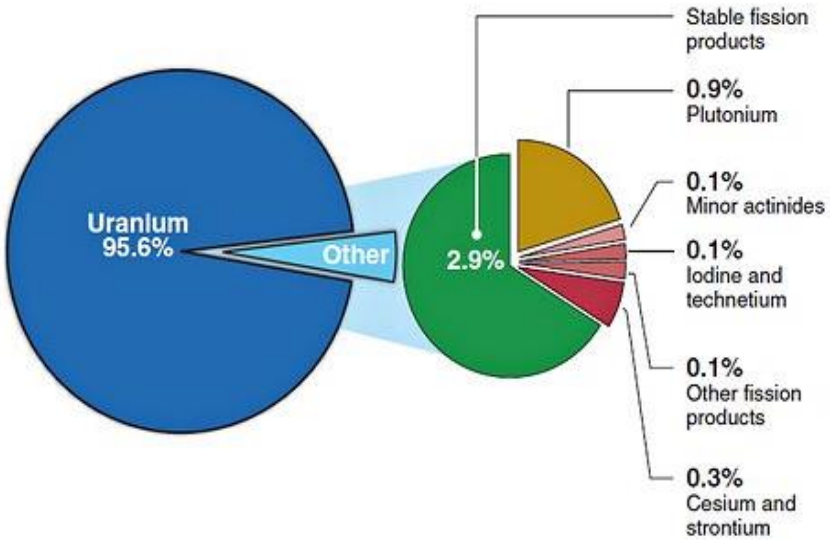
Burning of fuel

* T. Kaiba, Fig. 2, Advantages and dis-advantages of nuclear fuel reprocessing (2015)

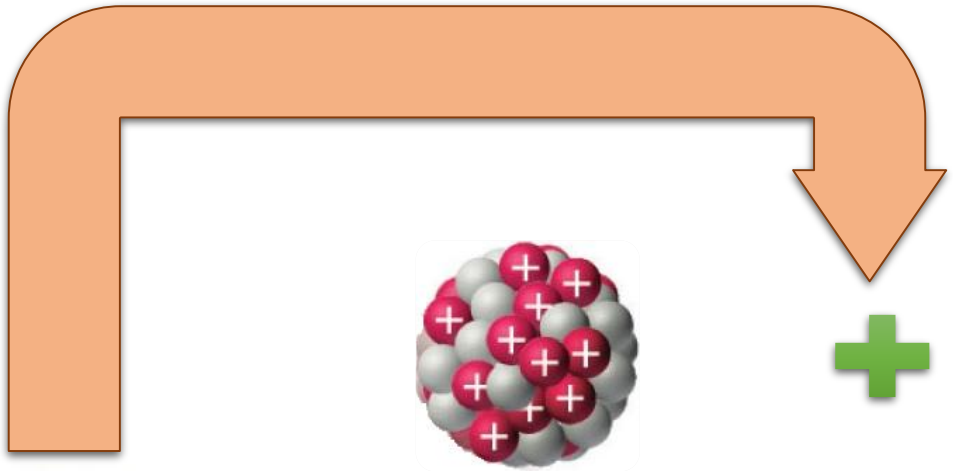
Nuclear Waste: A Dread



Nuclear reactor

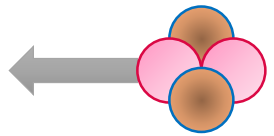


Spent Nuclear Fuel

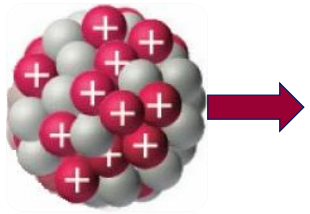


Fission fragments
(70-100 MeV)

Actinide Nucleus



α-particle
(4-6 MeV)



Recoil Nucleus
(70-100 keV)

* T. Kaiba, Fig. 2, Advantages and dis-advantages of nuclear fuel reprocessing (2015)

How can environment be saved from hazardous effects of radioactive wastes?

Waste Management

Nuclear reactor

Spent Nuclear Fuel

Open Fuel Cycle

Geological Disposal

PUREX & THOREX

Closed Fuel Cycle

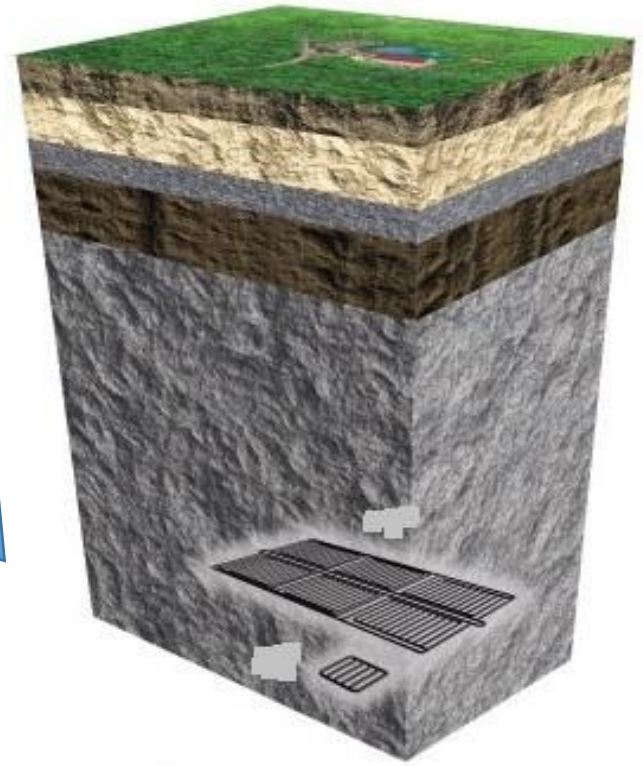
Reprocessing

High Level Wastes

Transmutation

Immobilization

Inert Matrix Fuel (IMF) Target



- Plants are-
  - ✓ Tarapur
  - ✓ Kalpakkam
  - ✓ Trombay

Potential Waste Forms

Glasses

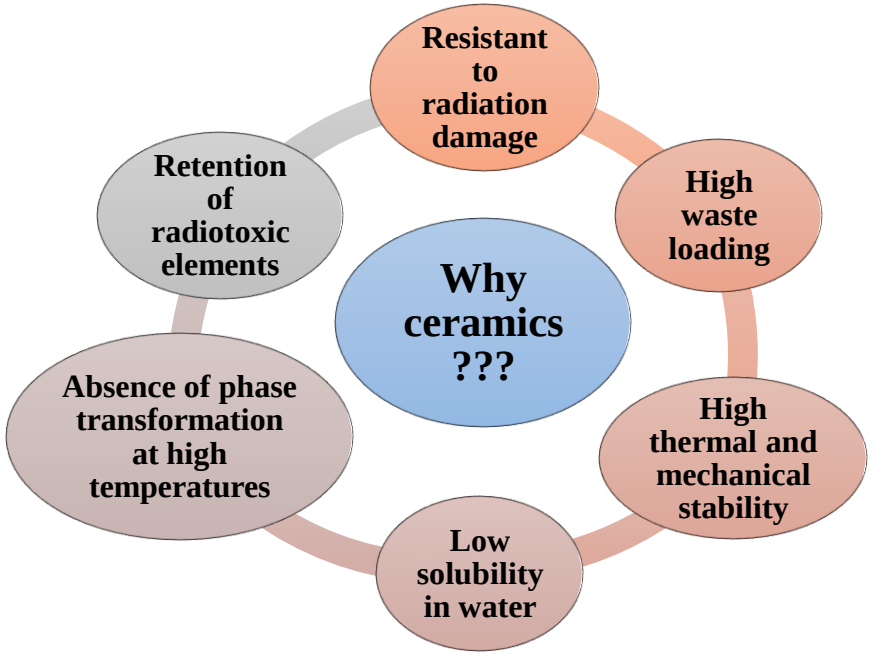
- Borosilicate glasses-- (Na-,Ln-, Pb-, Ca-)
- Alkali-Tin-Silicate glasses

Glass ceramics

- Phosphate glasses– (Fe-, Al-)

Ceramics

- Oxides-
  - Zirconia (ZrO_2)
  - Perovskite ($CaTiO_3$)
  - Pyrochlore ($A_2B_2O_7$)
  - Zirconolite ($CaZrTi_2O_7$)
  - Hollandite ($BaTi_8O_{16}$)
  - Spinel ($MgAl_2O_4$)
- Silicates-
  - Zircon ($ZrSiO_4$)
- Phosphates-
  - Monazite ($LnPO_4$)

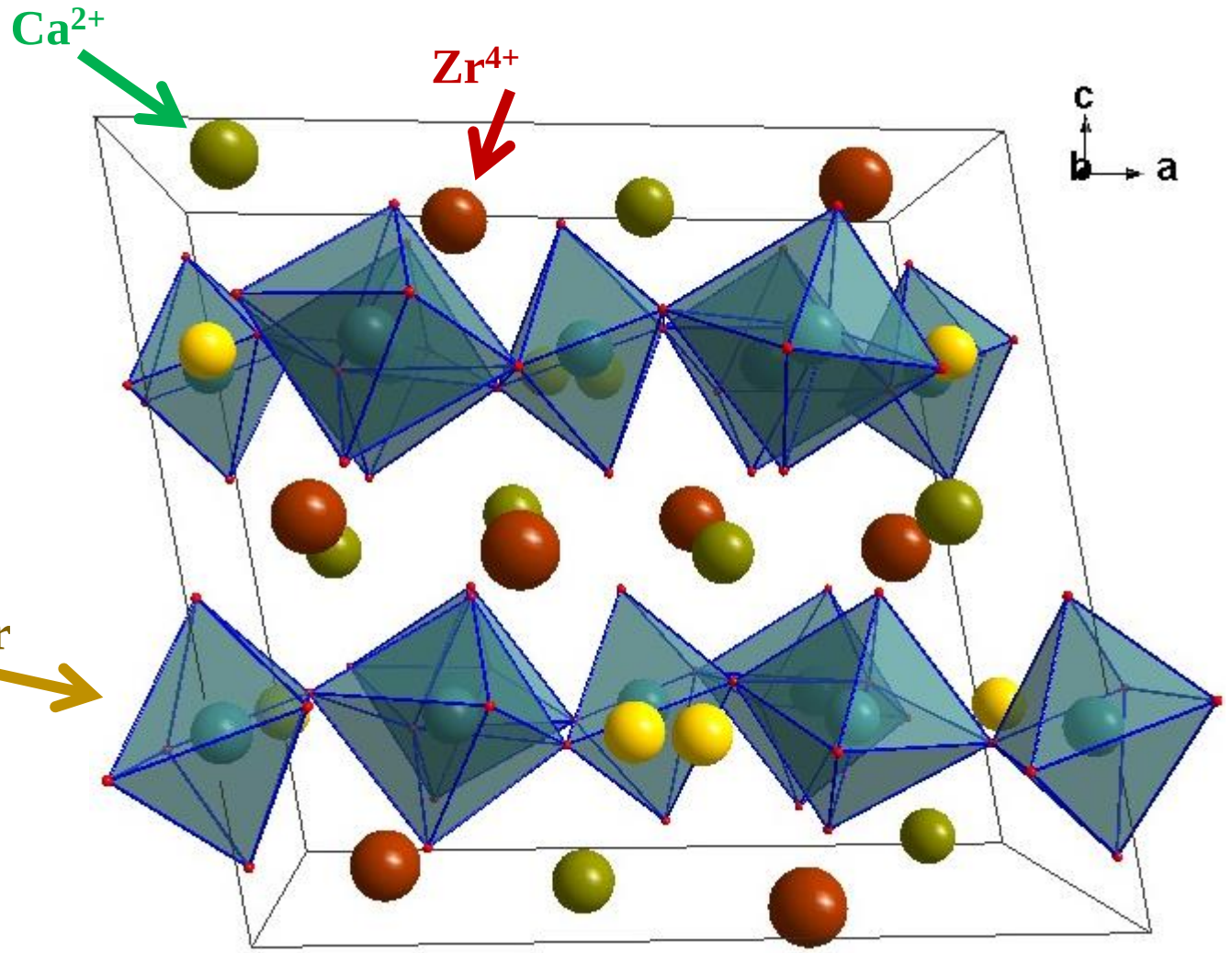


Preferable Ceramic Materials

S. No.	Materials	Aqueous Durability	Chemical Flexibility	Waste Loading	Radiation Tolerance	Volume Swelling
1.	Pyrochlore $Gd_2(Ti,Hf)_2O_7$	High	High	High	Low-High	Medium
2.	Zirconolite $CaZrTi_2O_7$	High	High	Medium	Low-Medium	Medium
3.	Zirconia $(Zr,Ln,Ac)O_{2-x}$	High	Medium	Medium	High	Low
4.	Zircon $ZrSiO_4$	High	Medium	Low	Low	High
5.	Monozite $LnPO_4$	High	Medium	High	High	Low
6.	Zirconates $Gd_2(Zr,Hf)_2O_7$	High	Medium	Medium	High	Low
7.	Perovskite $(Ca,Sr)TiO_3$	Low	Medium	Low	Medium	High

Zirconolite

- Zirconolite is one of the titanate based ceramic phase for the immobilization of actinides and lanthanides.
- Ideal chemical formula is $CaZrTi_2O_7$.
- It has a monoclinic layered type structure with space group $C_{2/c}$.

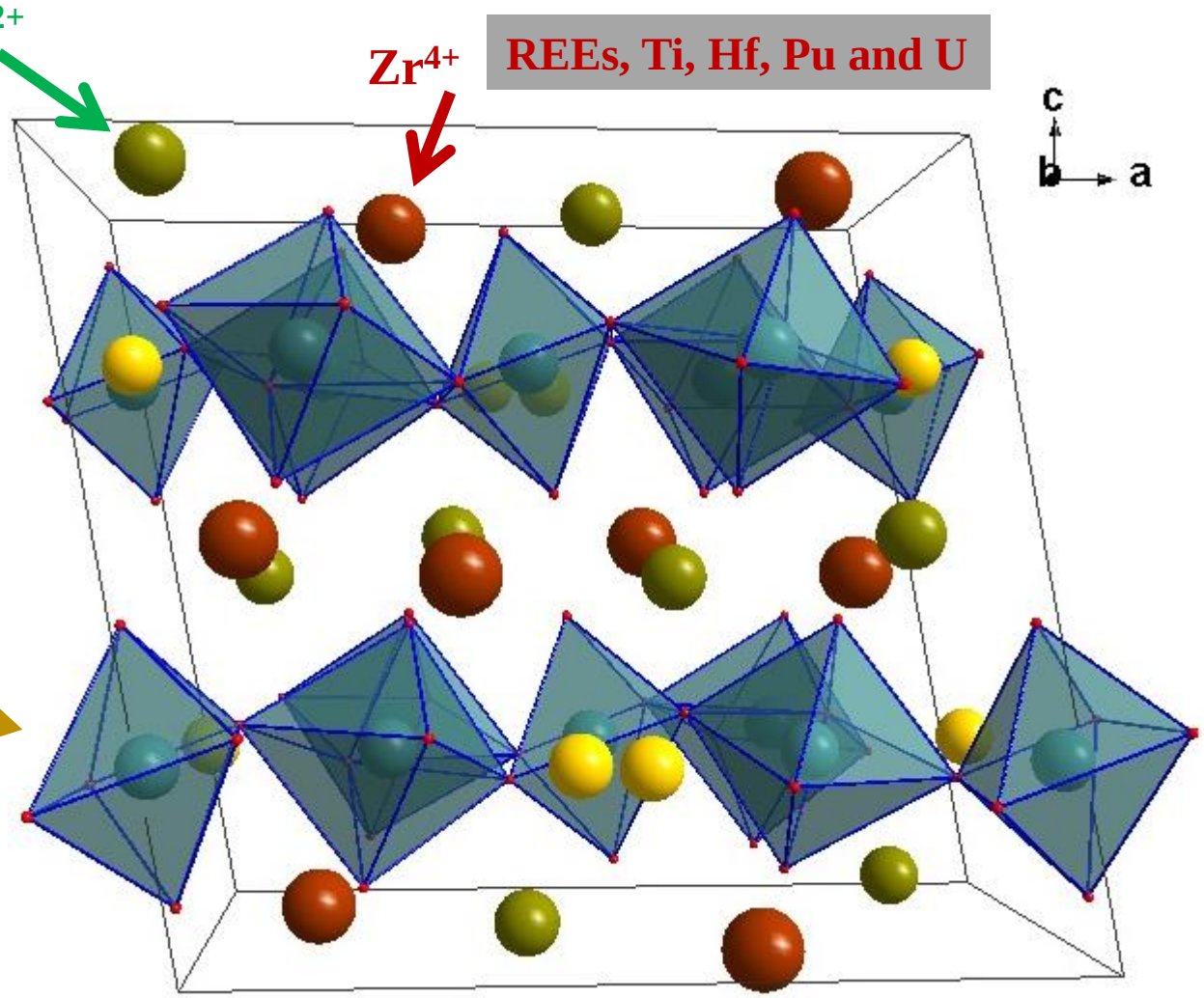


Zirconolite

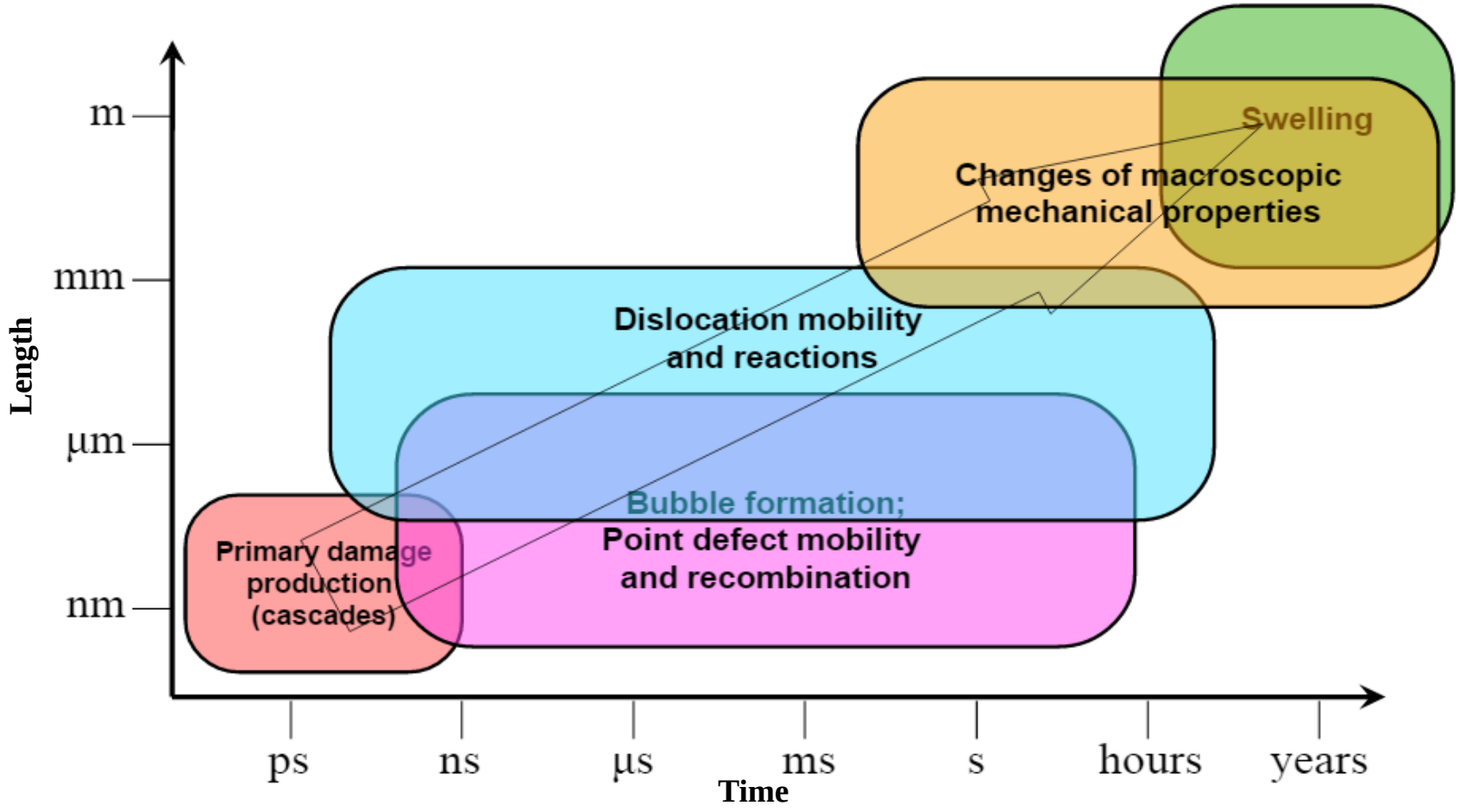
Rare-Earth Elements (REEs), Th, U, Mn and Sr

Waste Incorporation

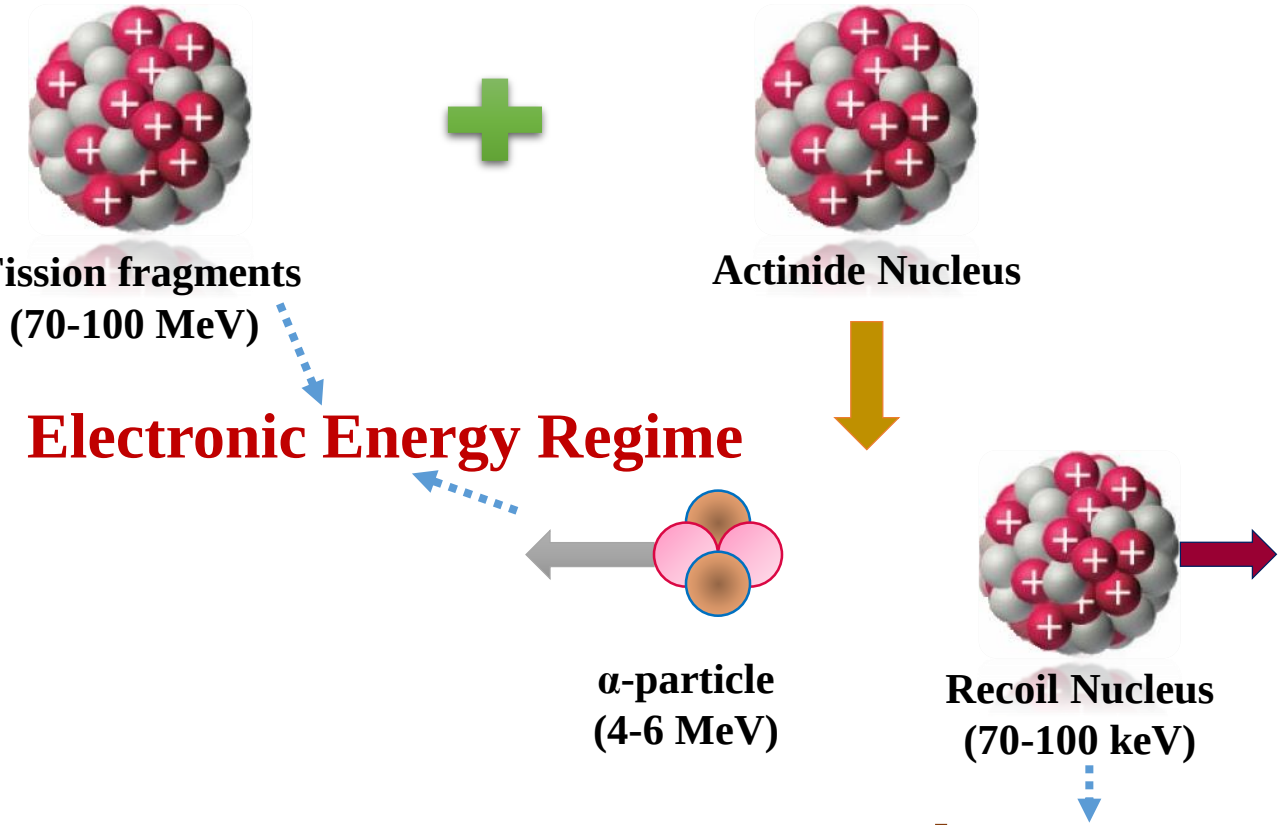
Mg, Fe (II), Fe (III), Nb, Pu, Al, Cr, Mn, Zn, Ta, W



Radiation Effects



Radiation Simulation

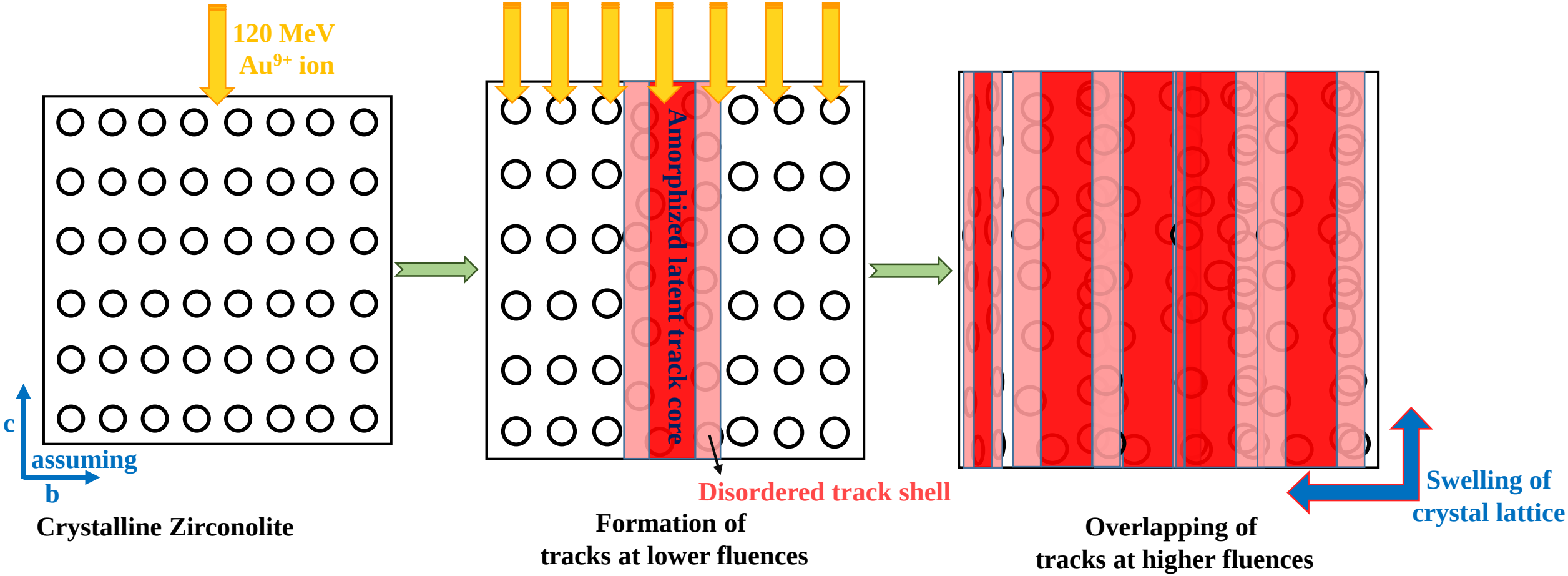


SRIM Calculations

Ion	Ion energy	S_e (keV/nm)	S_n (keV/nm)	S_e/S_n
Ba^+	70 MeV	13.47	0.202	66.7
Au^+	120 MeV	21.67	0.325	66.7

Swift Heavy Ion Induced Effects

Thermal Spike Model



Crystalline Zirconolite

Formation of tracks at lower fluences

Overlapping of tracks at higher fluences

Research Activities

Synthesis

Solid State Reaction

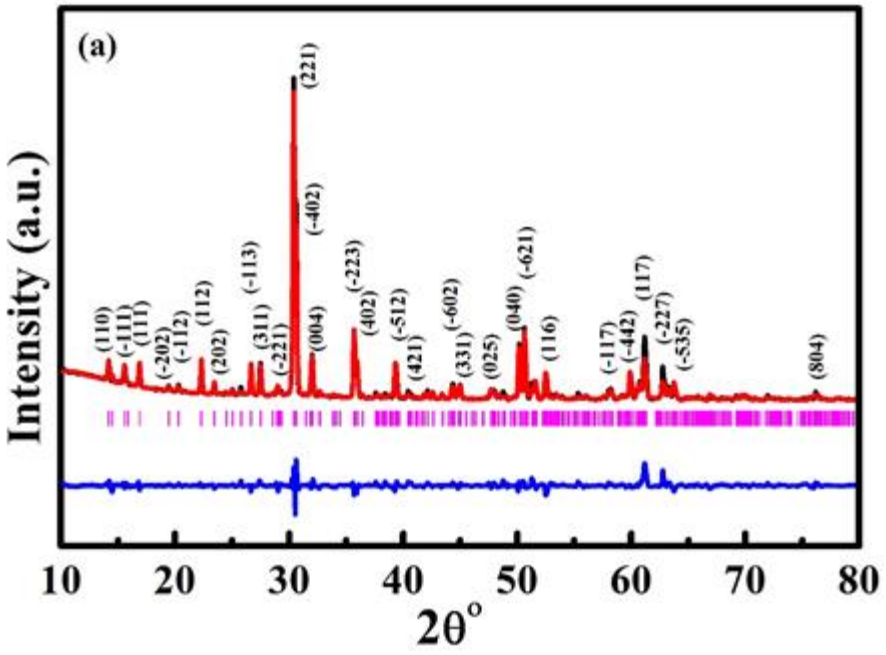
$$CaCO_3 + Nd_2O_3 + ZrO_2 + 2\,TiO_2 + Al_2O_3 \rightarrow Ca_{0.8}Nd_{0.2}ZrTi_{1.8}Al_{0.2}O_7 + CO_2$$

- Homogenization and pelletization
- Sintering: 1200 ^oC/4 hrs and 1400 ^oC/16 hrs
- Heating rate: 3 ^oC/min
- Cooling rate: 2 ^oC/min

Research Activities

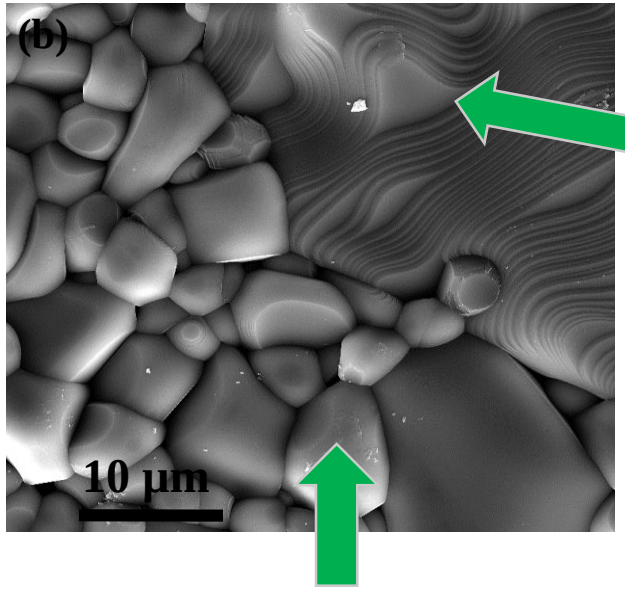
Characterization

XRD



- Monoclinic structure with space group $C_{2/c}$
  a = 12.4526(5) Å, b = 7.2694(3) Å,
  c = 11.3844(4) Å and β = 100.605(3)°

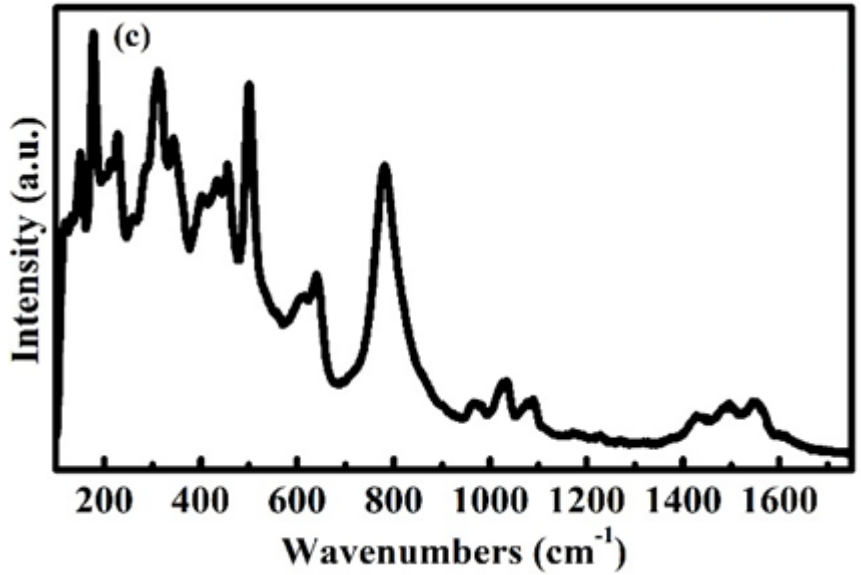
SEM



Lamellar Structure

Grain size ~7 µm

Raman

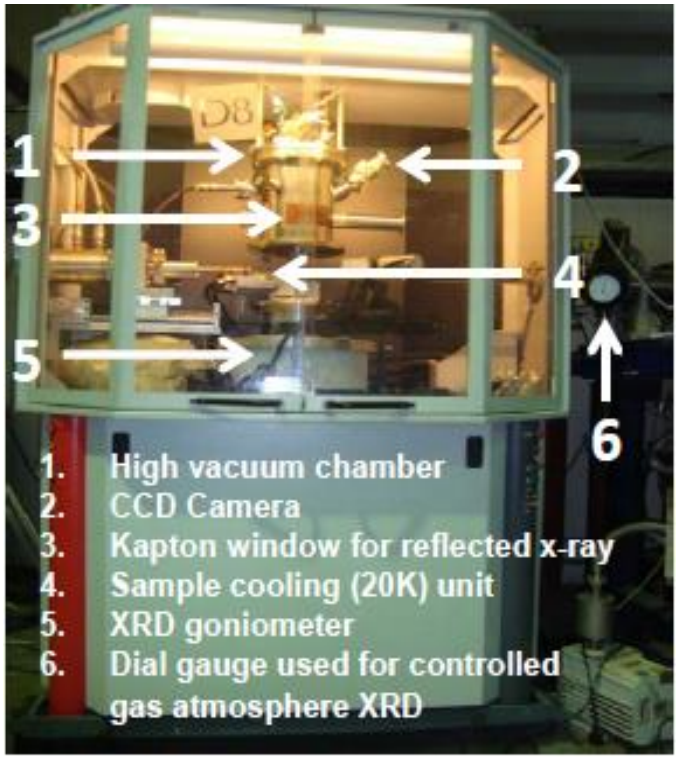


*M. Gupta *et. al.* Nucl. Instr. Meth. B 379 (2016) 119

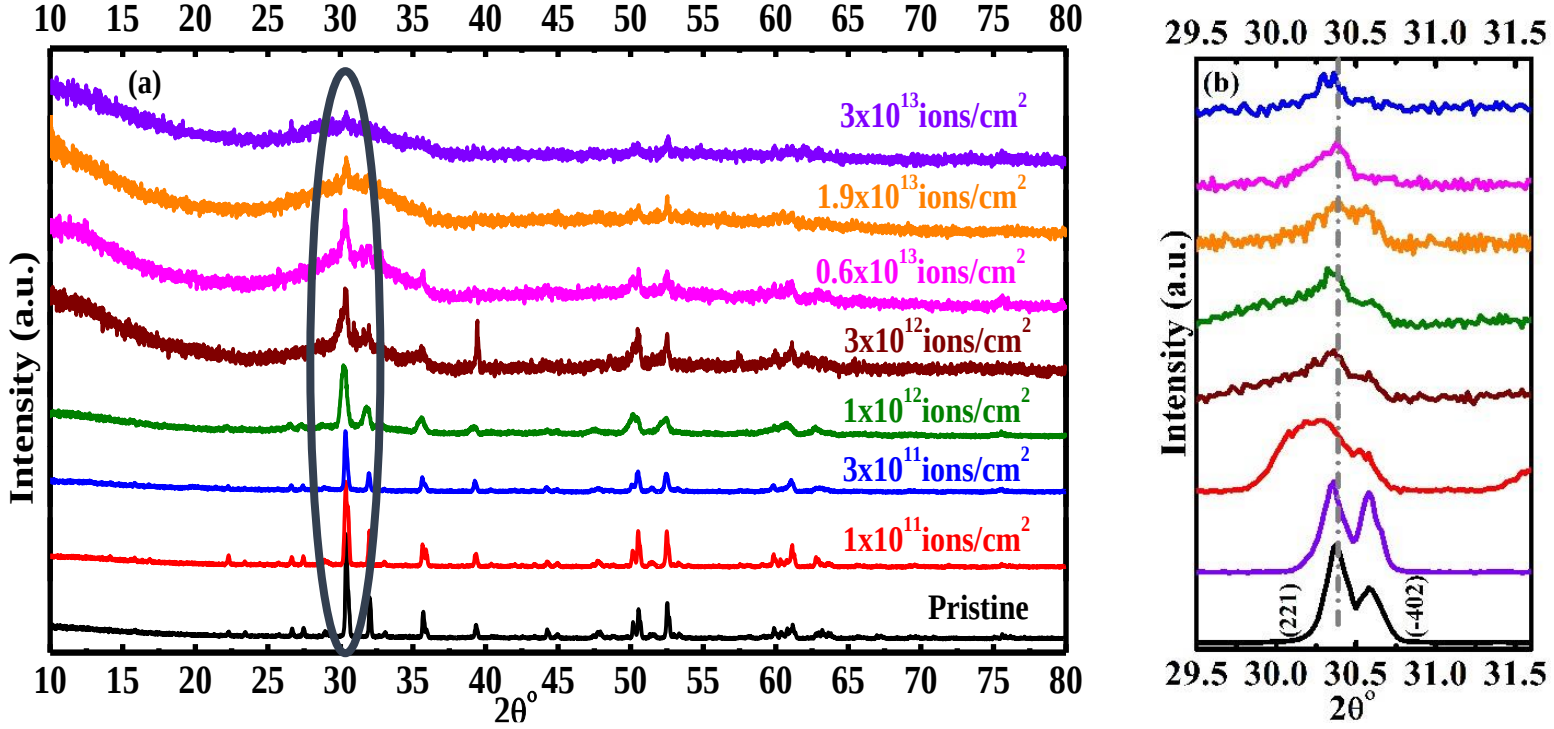
Research Activities

Irradiation with 120 MeV Au^{+} Ions

****In-situ* XRD facility in LINAC beamline using 16 UD Pelletron Accelerator at IUAC Delhi**



Irradiation Induced XRD Effects

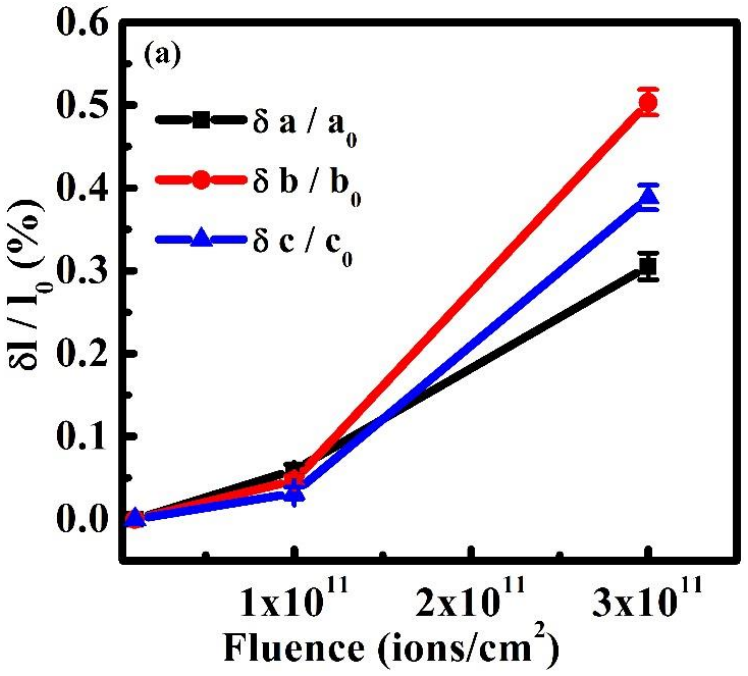


*Kulriya *et. al.*, Rev. Sci. Instrument 78 (2007) 113901

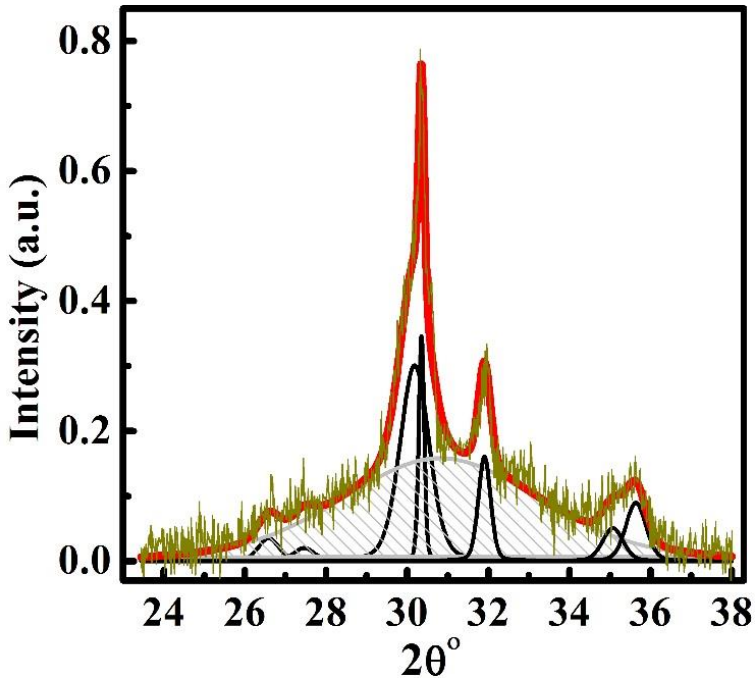
Research Activities

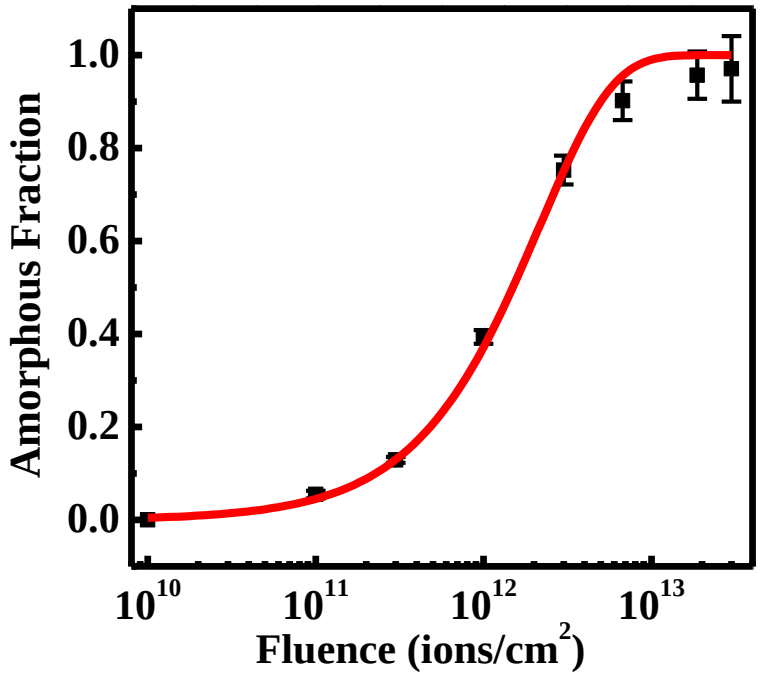
Irradiation Induced XRD Effects

Lattice Expansion



Amorphous Fraction Calculation





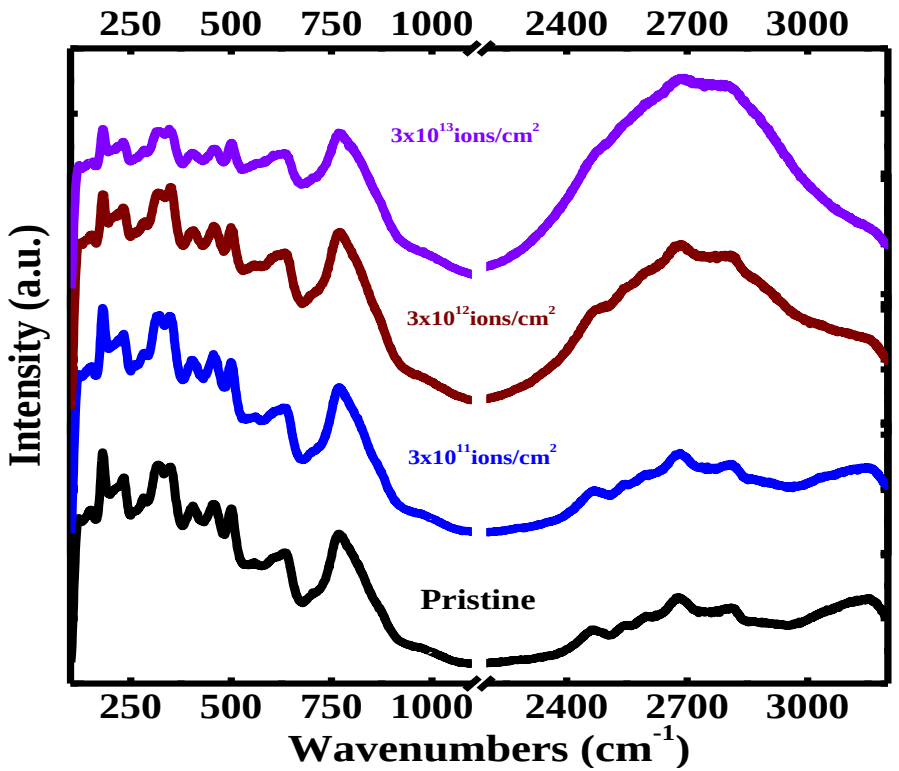
Area cross-section, σ - 69.6 ± 4 nm^2

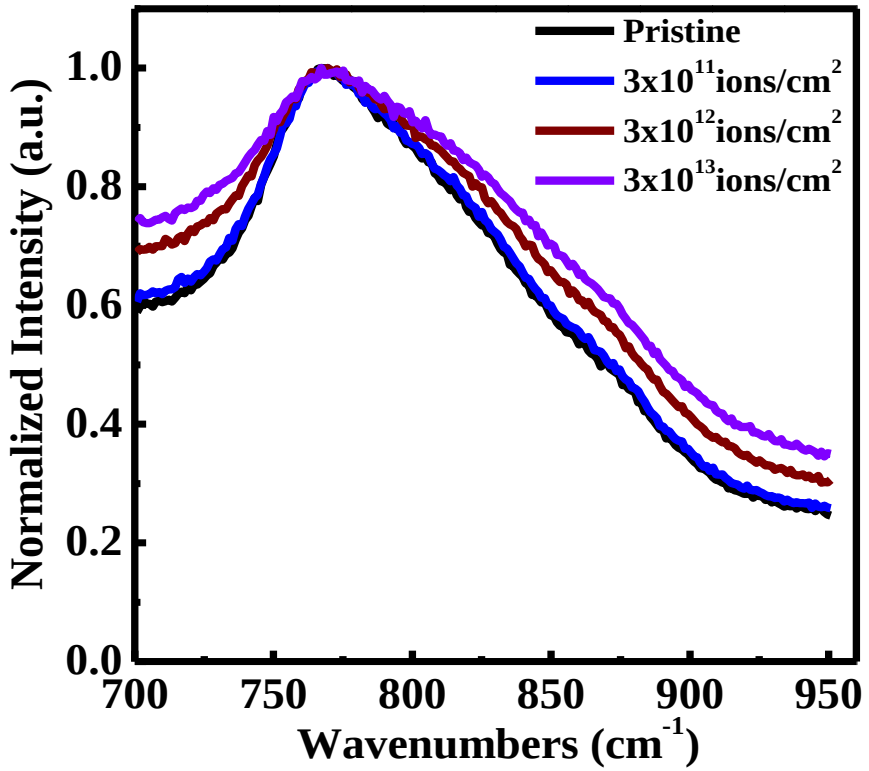
Ion track diameter, d - 9.4 ± 0.3 nm

$$f_a(\phi) = A\,(1 - e^{-\sigma\phi})$$

Research Activities

Irradiation Induced Raman Effects





Summary

- Irradiation with 120 MeV Au^{+} ions in fluence range of $1\times10^{11\text{-}14}$ ions/cm^2 at room temperature

- XRD and Raman investigations:
  - Loss of crystallinity
  - Amorphization in the form of ion tracks
  - Bonds distortion to TiO_5

- Stability and durability of zirconolite in high (electronic) energy regime too

Acknowledgements

- IUAC Pelletron Group, IUAC facilities, New Delhi

Thank You!!!