Spatial profile studies

of

Nuclear material burn-up

by

Laser Ablation- Ionization

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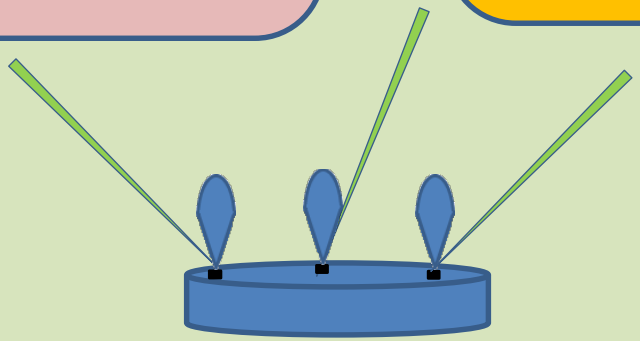
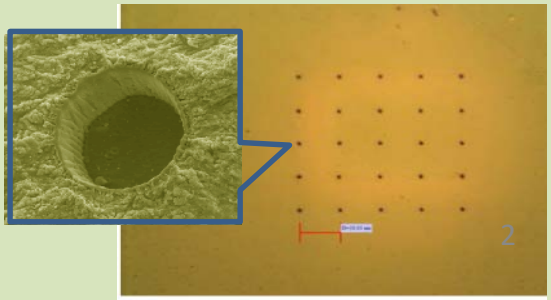
Burn-up measurements

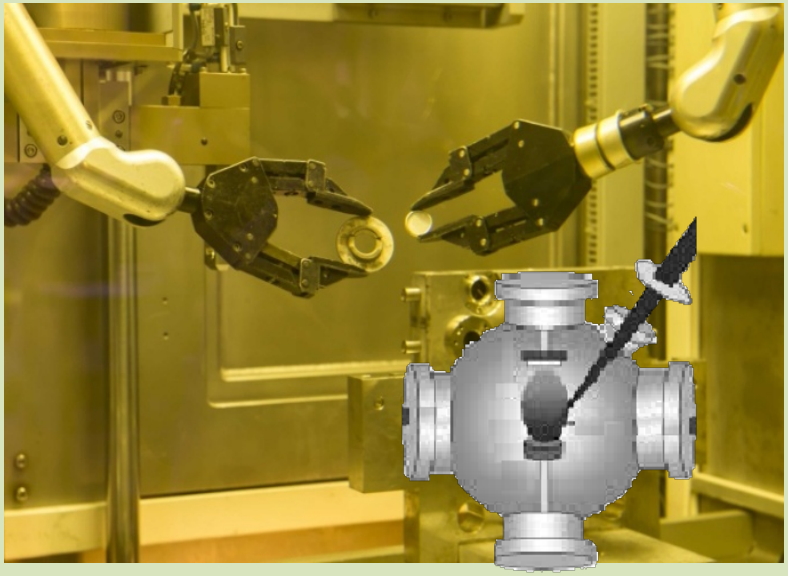
Laser as Sampling Tool

Chemical Dissolution Method	Proposed Laser Ablation
➢ Dissolution ➢ Chemical Separation ➢ Mass Spectrometer Analysis	➢ Avoids Dissolution ➢ Avoids Chemical Separation ➢ Mass Spectrometer Analysis
❑ Traditional, ASTM Prescribed ❑ Spatial Profiling not possible ❑ Time consuming & Labour intensive ❑ Sample to be brought to laboratory	❑ New ❑ Spatial Profiling is possible ❑ Quick & Direct sampling ❑ Laser - taken inside Hot Cells





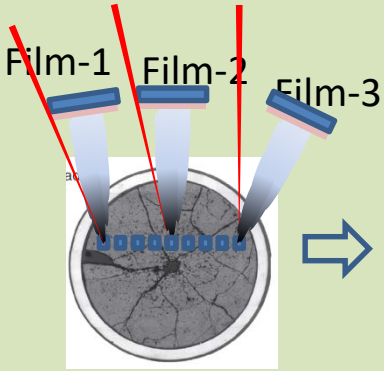
Pulsed Laser -A Tool for analytical sampling



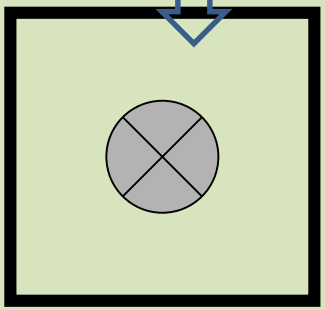
Method -1

Deposit Thin films inside Hot Cell

Dissolve films in nitric acid

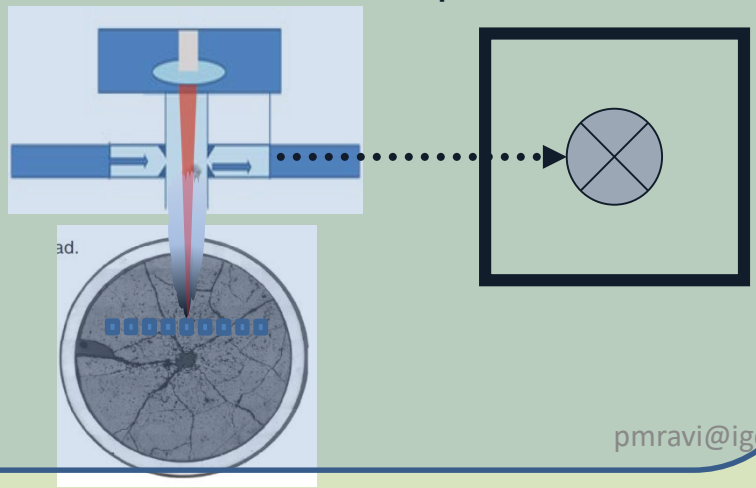


Analyze dissolved films in mass spectrometer



Method -2

Directly feed Laser ablated plume to mass spectrometer



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Laser Ablation Process

High Power Nanosecond Laser Pulse

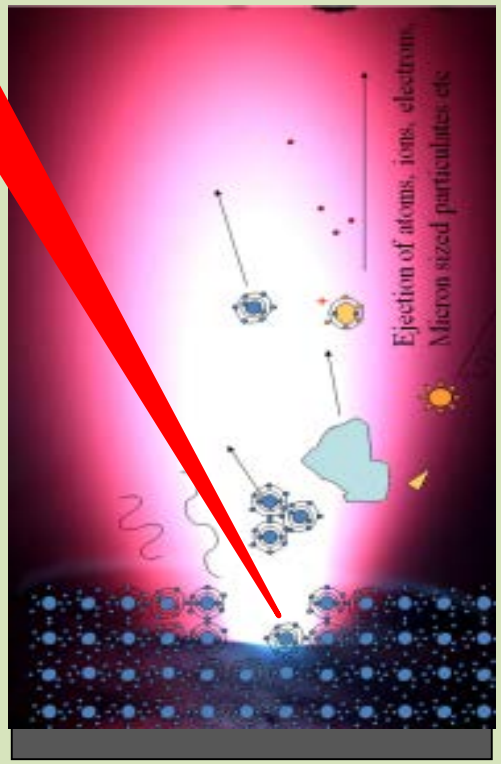
- Laser Pulse strike material surface
- Thermal & non-thermal transfer of energy
- Following Physical Process Occurs

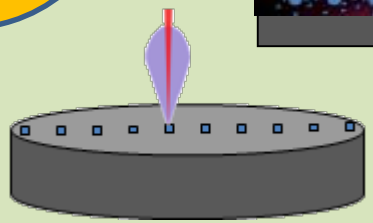
(i) Evaporation (Laser heating)

(ii) Electron & Ion ejection

(iii) Particulates & atomic clusters ejection

(iv) Plasma light emission





Laser Ablation Process

Physical Process of Laser Ablation

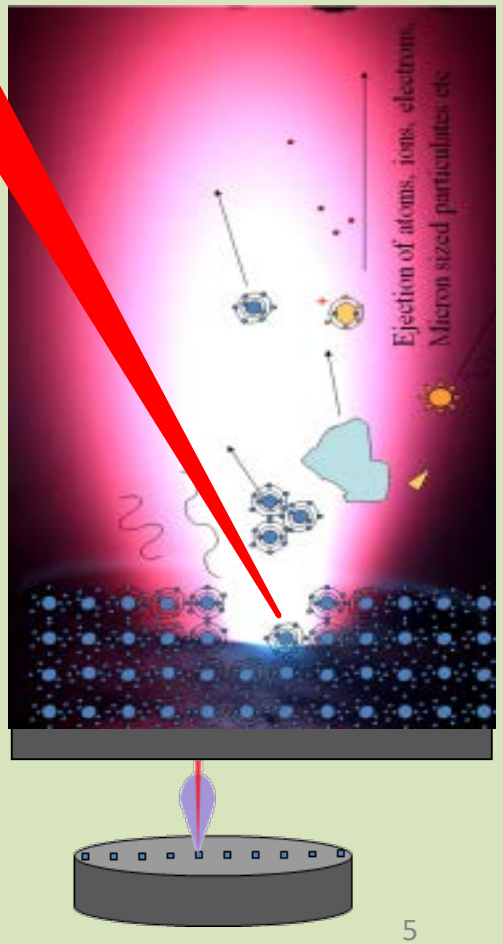
(i) **Evaporation (Thermal Process - Heating)**

- Neutral atom & molecules ejection.
- Vapour composition governed by partial pressure of vaporizing species.
- Temperature 3000 – 7000K (ns pulsed laser heating of refractory materials).

(ii) Electron & Ion ejection

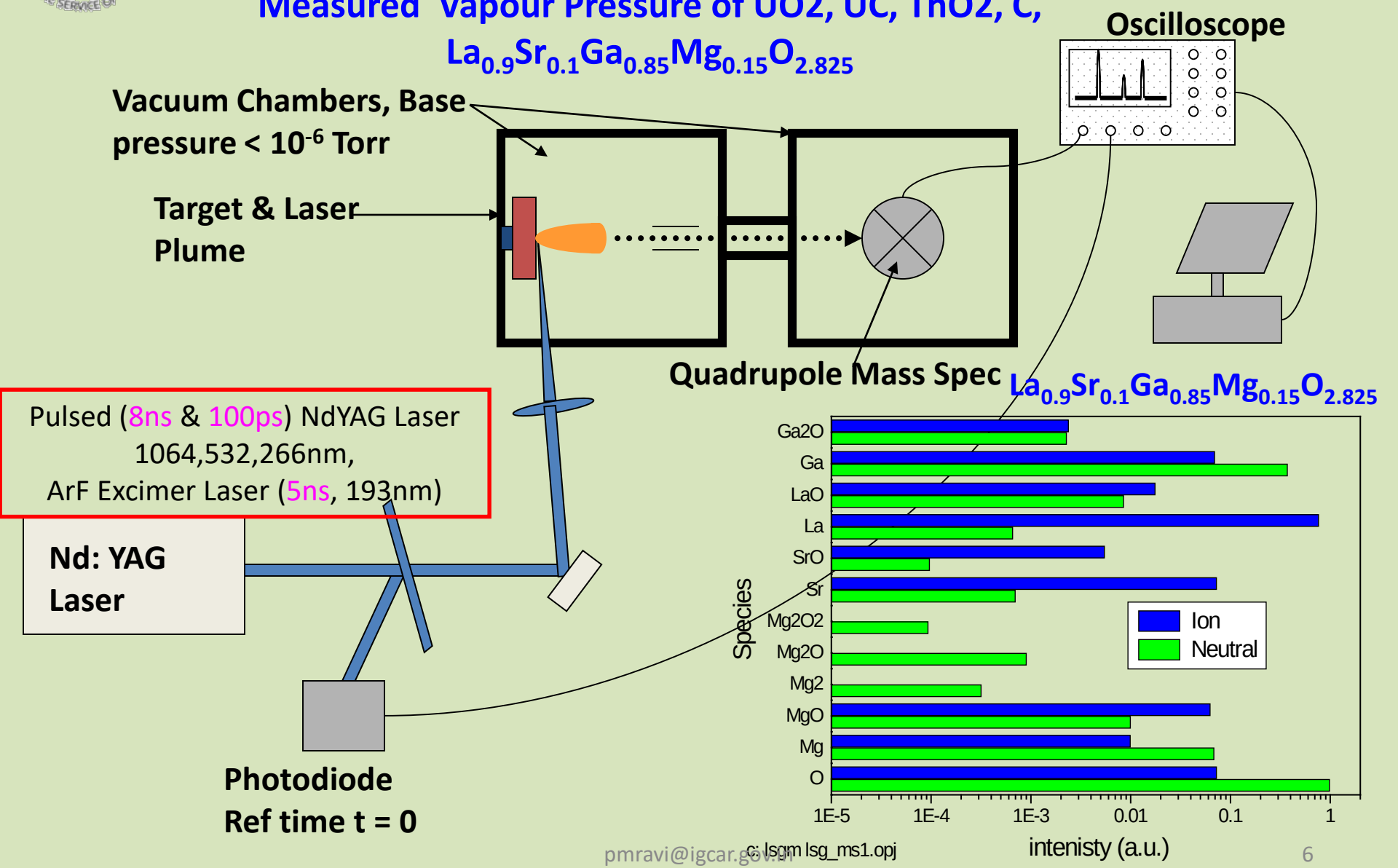
(iii) Particulates & atomic clusters ejection

(iv) Plasma light emission



Laser Vapourization -Mass Spectrometer (using QMS)

Measured Vapour Pressure of UO2, UC, ThO2, C, $La_{0.9}Sr_{0.1}Ga_{0.85}Mg_{0.15}O_{2.825}$



Laser Ablation Process

Physical Process of Laser Ablation

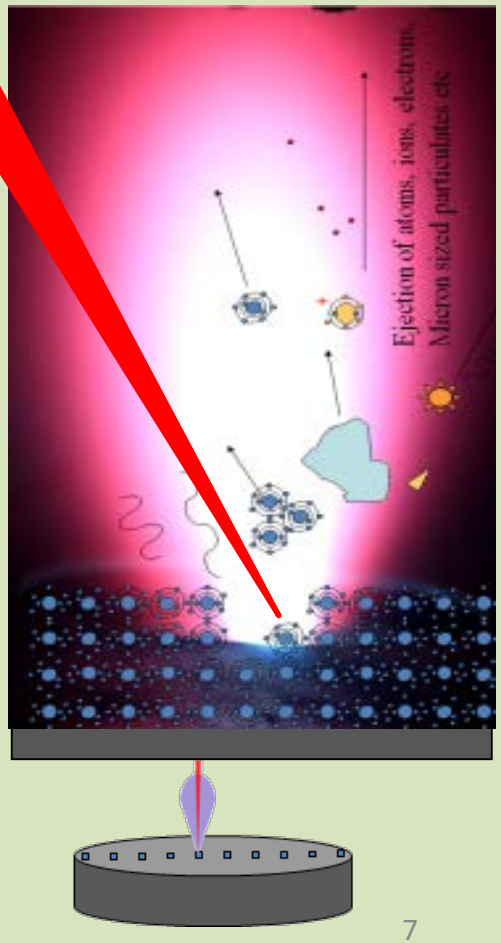
(i) Evaporation (Thermal Process - Heating)

(ii) Electron & Ion ejection

- Laser produced Ions are used.
- Time of Flight Mass Spectrometer.
- Spatial profiling / surface chemical & isotope mapping.
- Quick Burn-up measurements.

(iii) Particulates & atomic clusters ejection

(iv) Plasma light emission



Laser ablation – Time of flight Mass spectrometer

^{10}B Isotope measurement in Irradiated B_4C

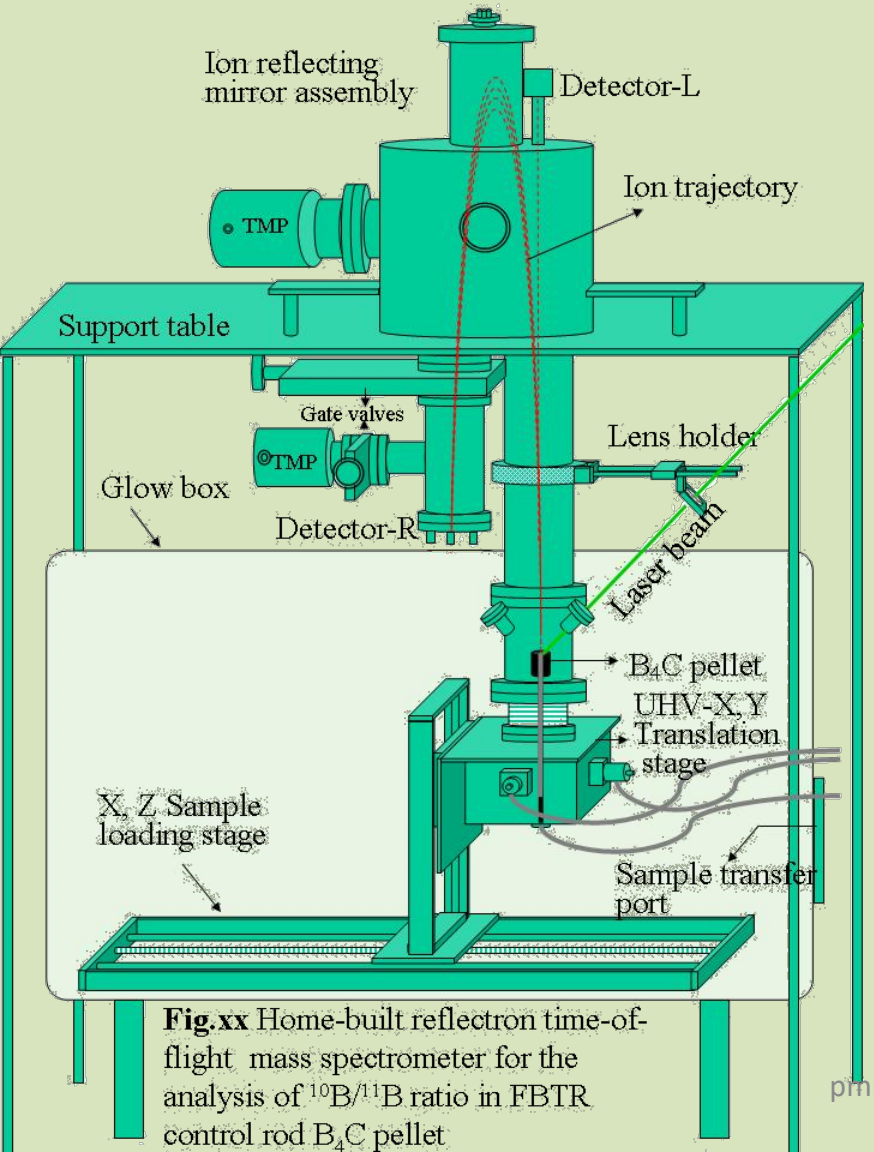
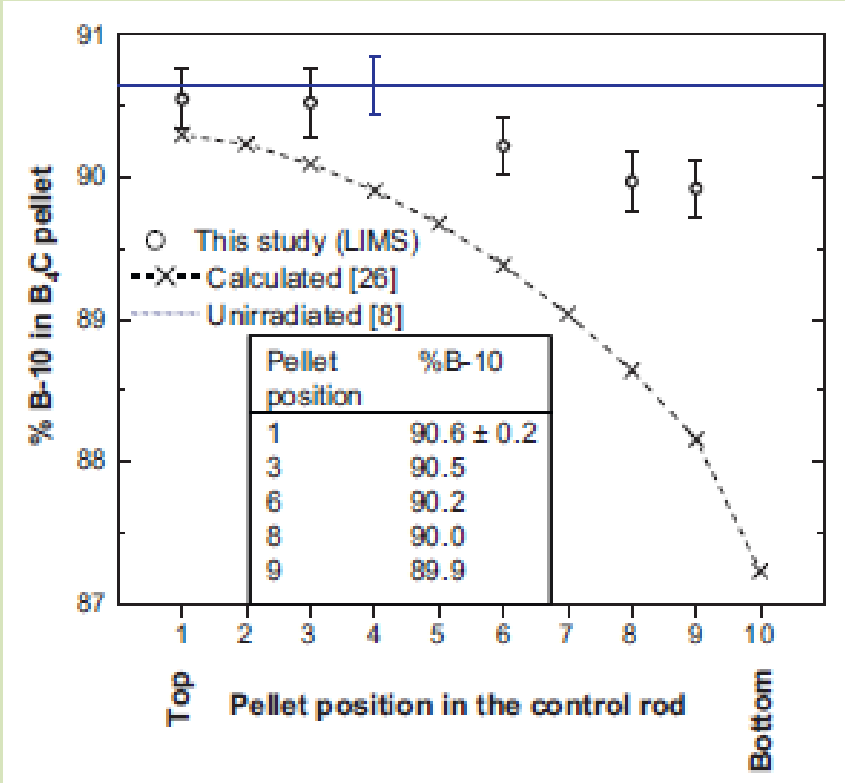
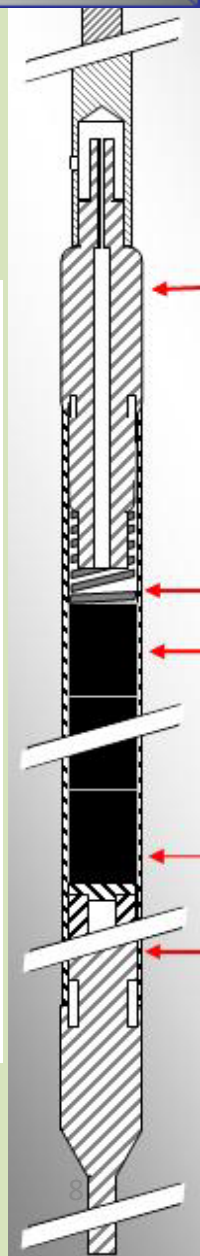


Fig.xx Home-built reflectron time-of-flight mass spectrometer for the analysis of $^{10}B/^{11}B$ ratio in FBTR control rod B_4C pellet

$^{10}B/^{11}B$ = 0.25 (natural)

$^{10}B/^{11}B$ = 0.9 (FBTR control rod)





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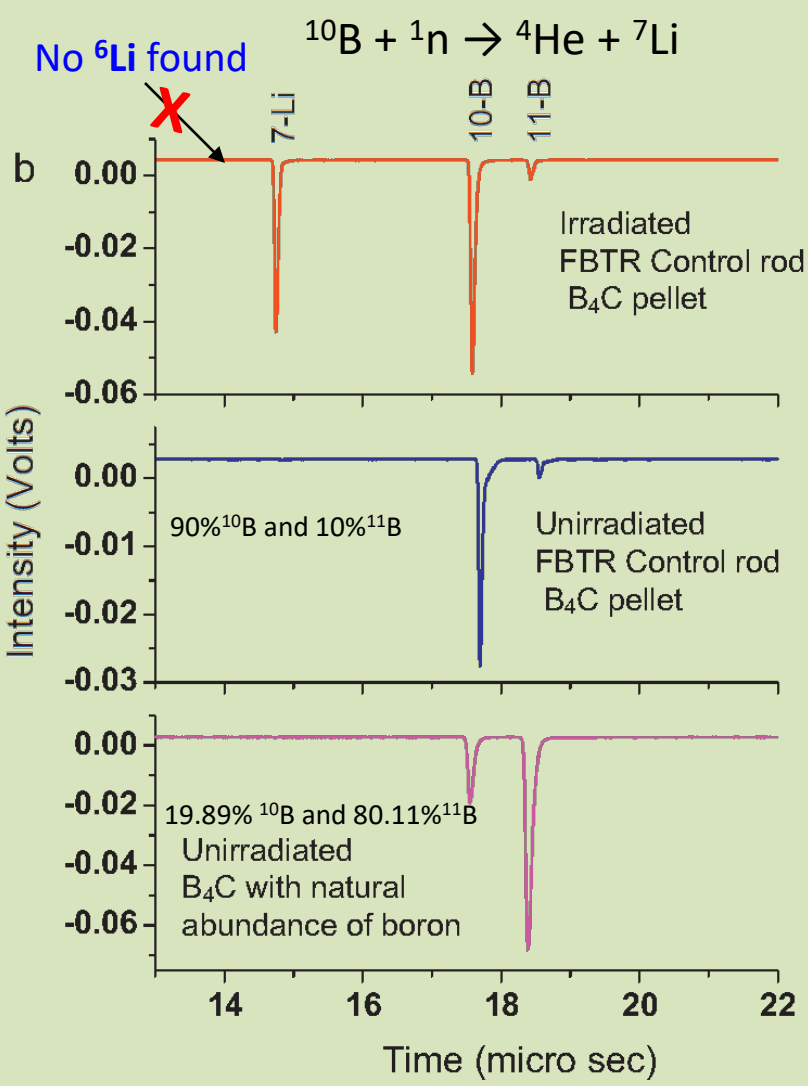
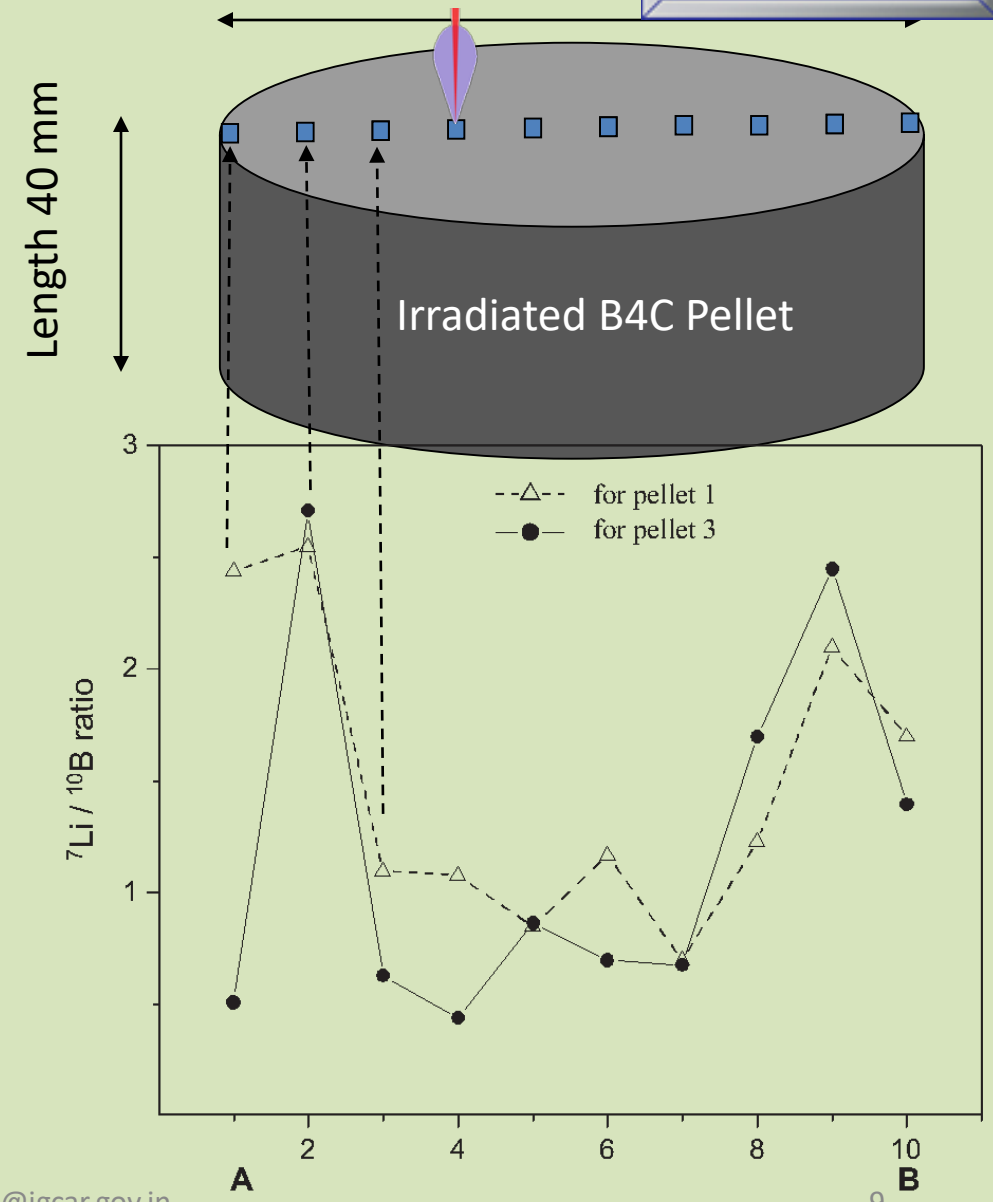
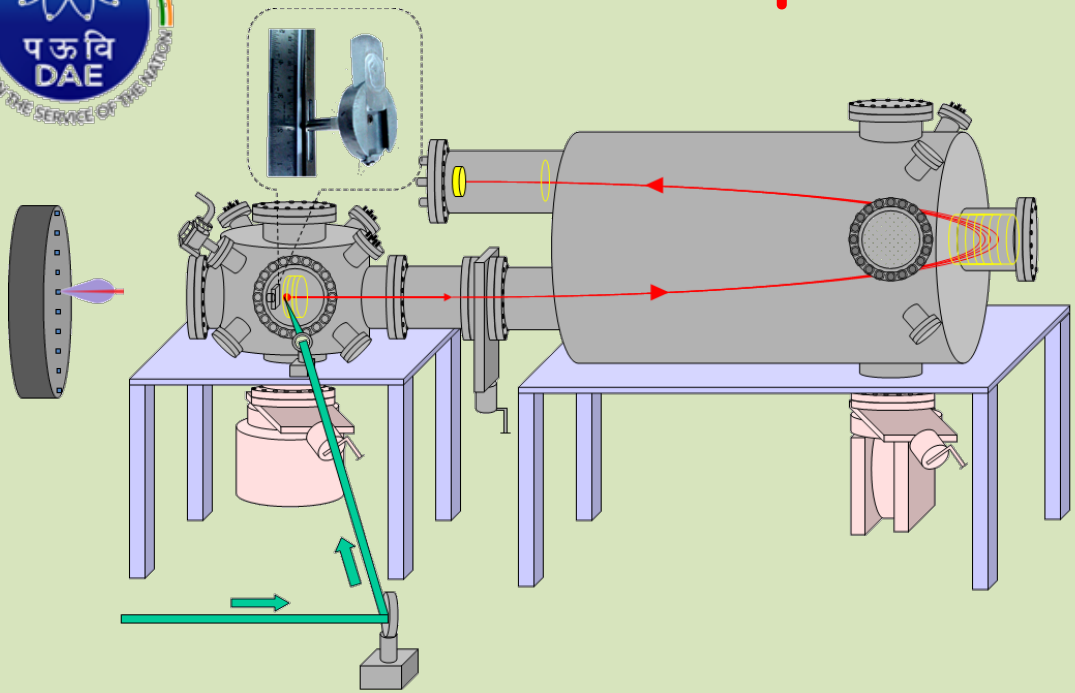
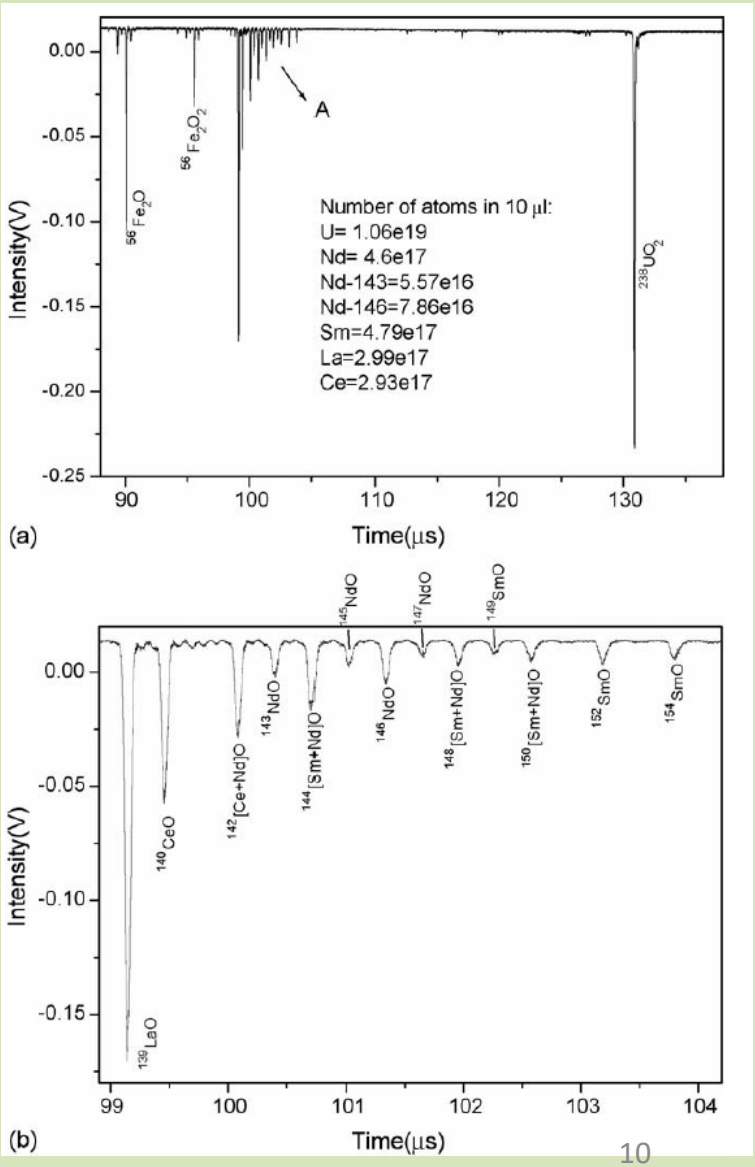
Spatial Profile of ^{10}B Isotope consumption in Irradiated B_4C

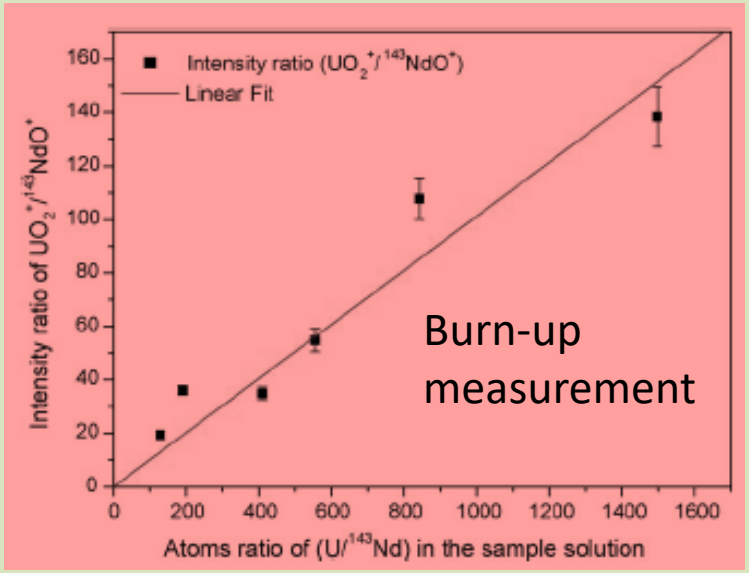




Laser-Mass Spectrometer (TOF)







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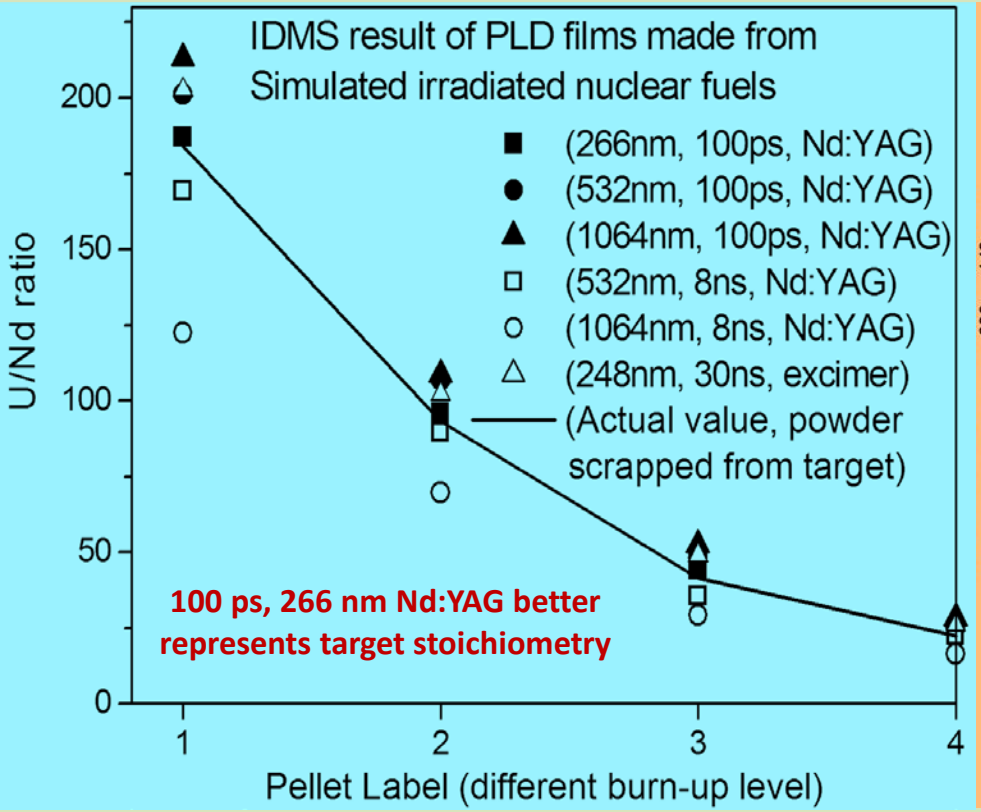
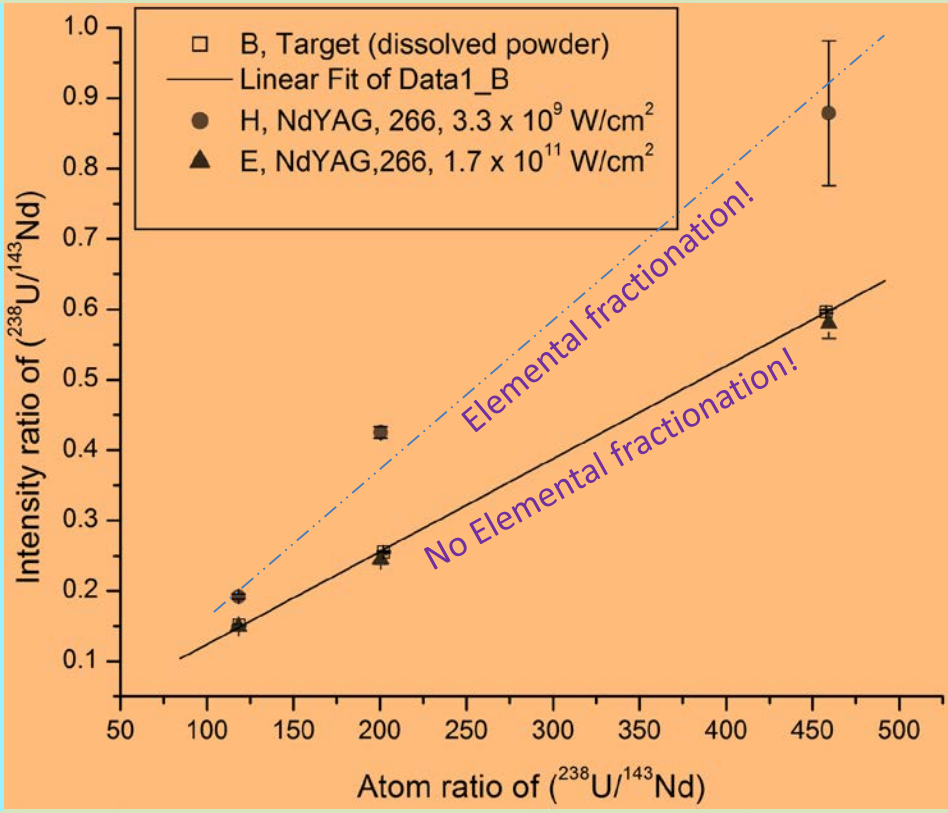
Which laser?



Lower Wavelength & Lower Pulse duration are better!

At What Fluence?



Higher fluence is better!
However, Very high fluence is bad

Laser Ablation Process

Physical Process of Laser Ablation

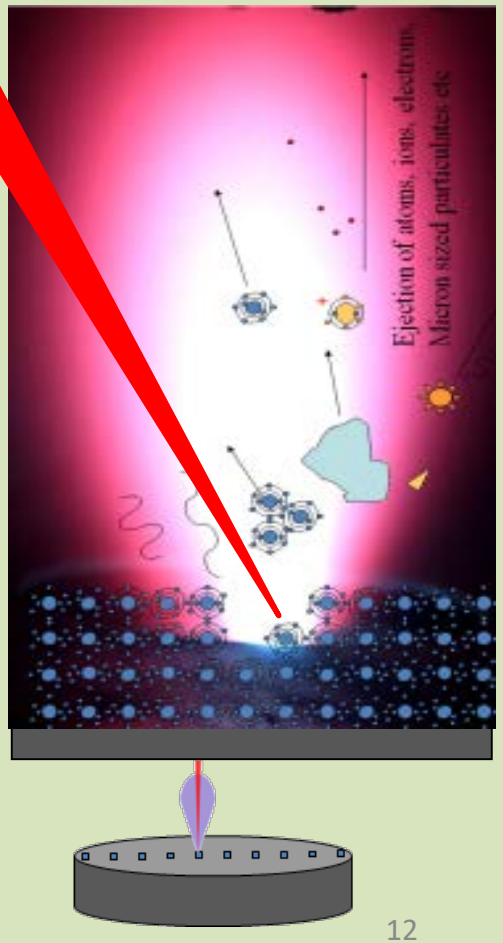
(i) Evaporation (Thermal Process - Heating)

(ii) Electron & Ion ejection

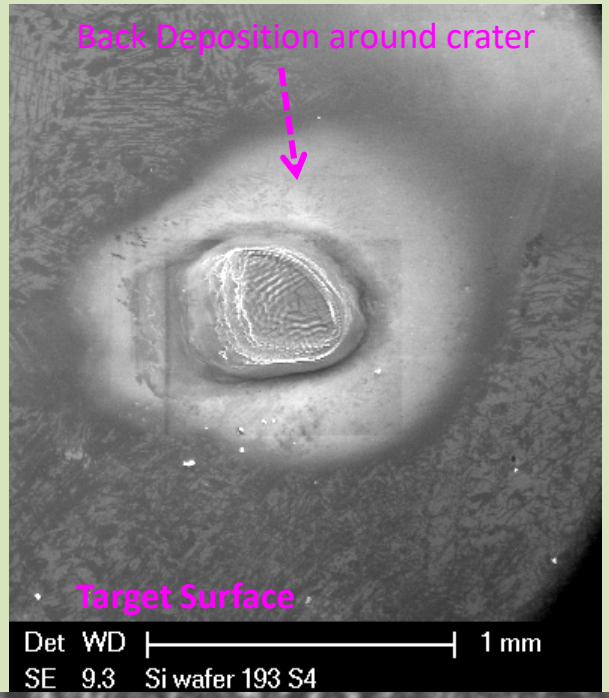
(iii) Particulates & atomic clusters ejection

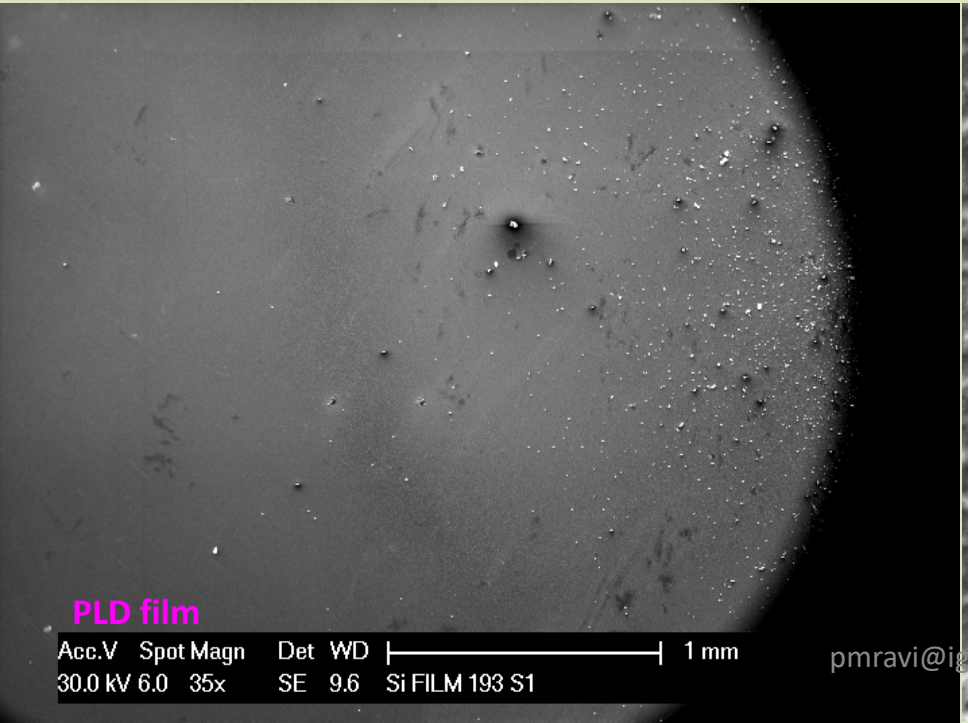
- Chip Off (broken from surface)
- Sub-surface boiling-condensation
- Sub micron size
- Ensures exact target chemical composition
- Perfect for ICP plasma digestion
- Sampling input for ICPMS ➔ LA-ICPMS
- Spatial Profiling / Chemical & isotope mapping

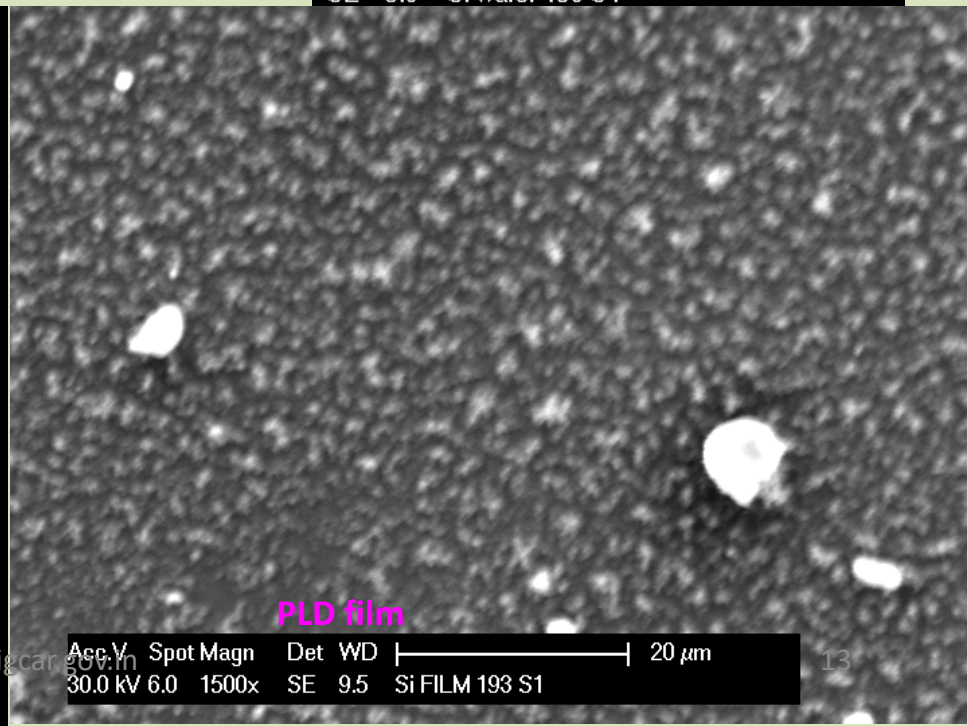
(iv) Plasma light emission



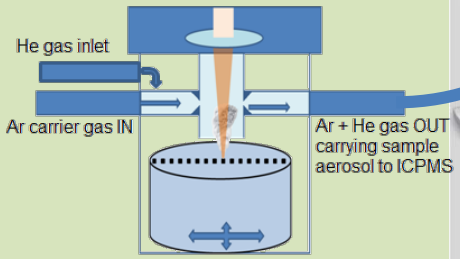
- Vapour Back Deposition (ambient pressure).
- Particulates: not back deposited.
- Particulates around 0.5 micron size – good for ICP Plasma “digestion”.
- Low laser fluence favours vapour.
- High laser fluence promotes particulates.

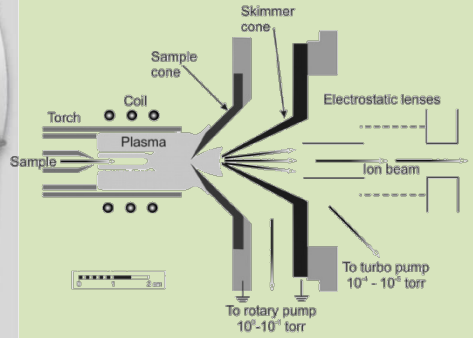


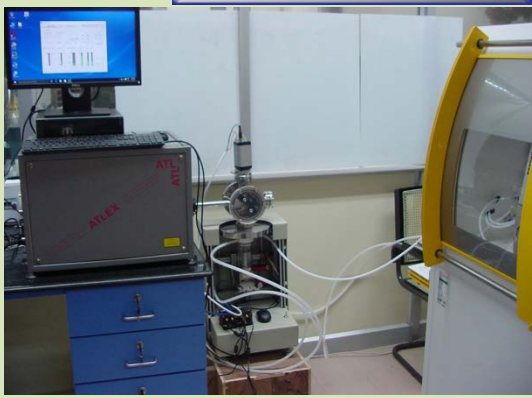




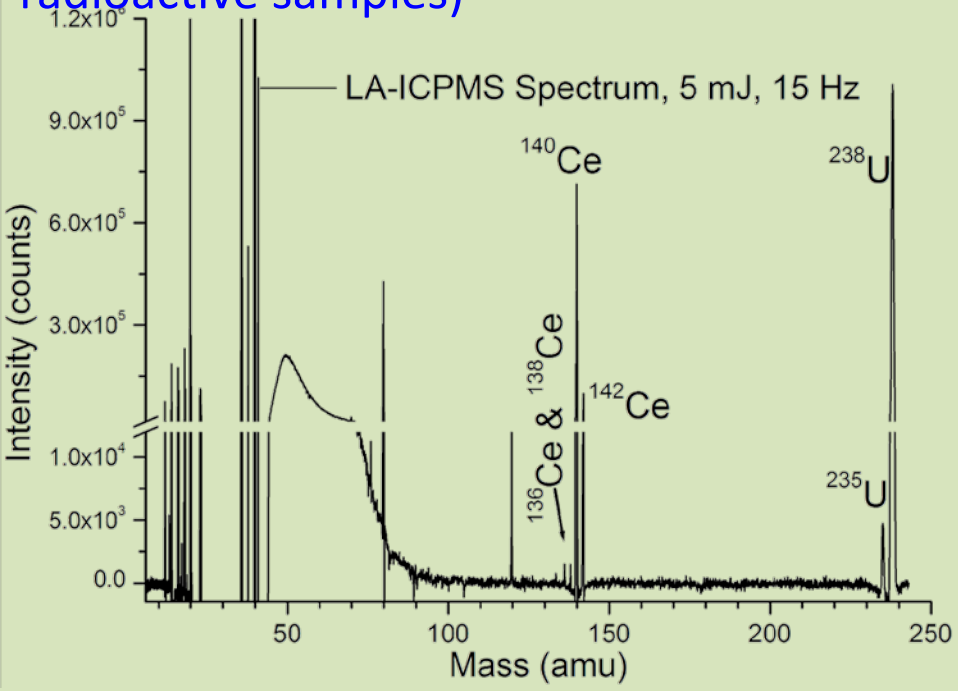
Laser Ablation-ICPMS

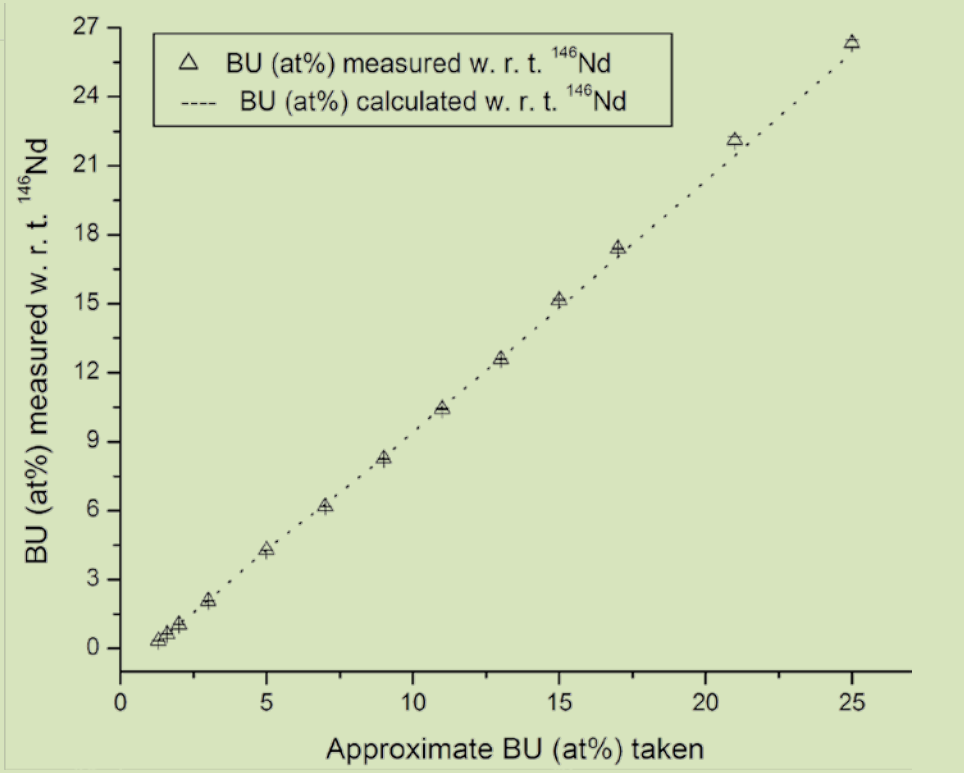






Home designed LA cell (suitable for radioactive samples)





Laser Ablation Process

Physical Process of Laser Ablation

(i) Evaporation (Thermal Process - Heating)

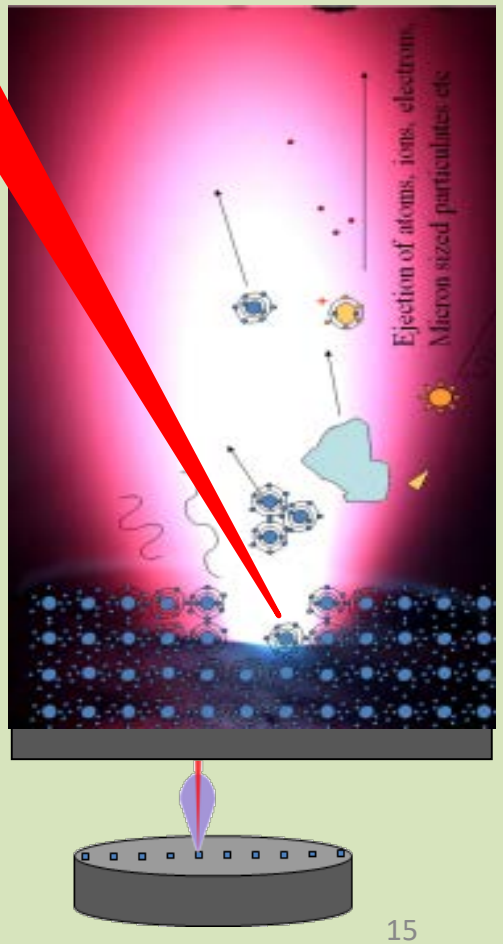
(ii) Electron & Ion ejection

(iii) Particulates & atomic clusters ejection

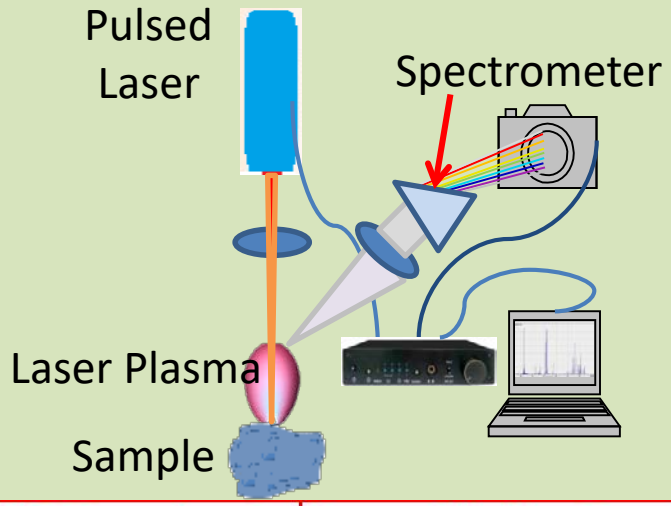
Mass Spectroscopy

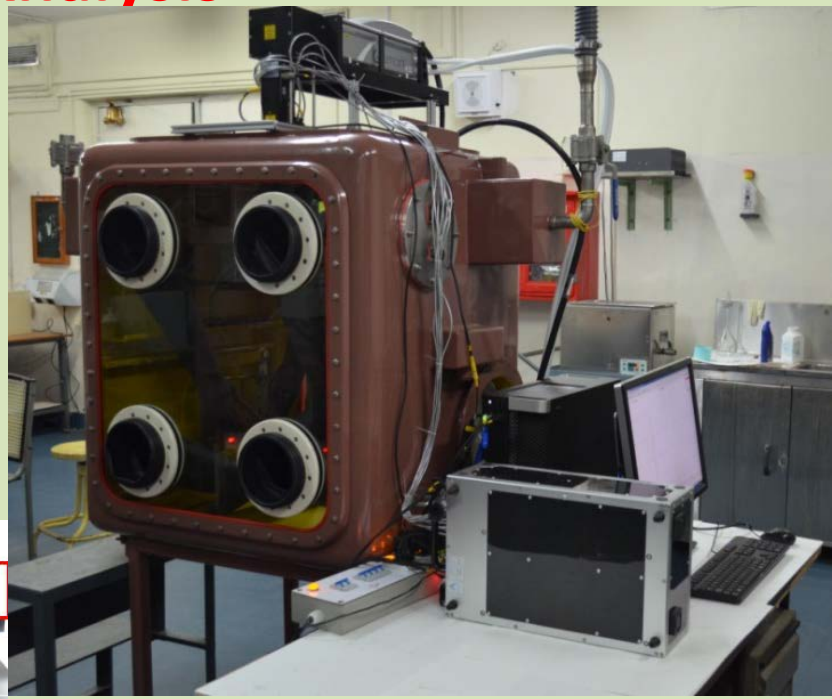
(iv) Plasma light emission (*Optical Spectroscopy*)

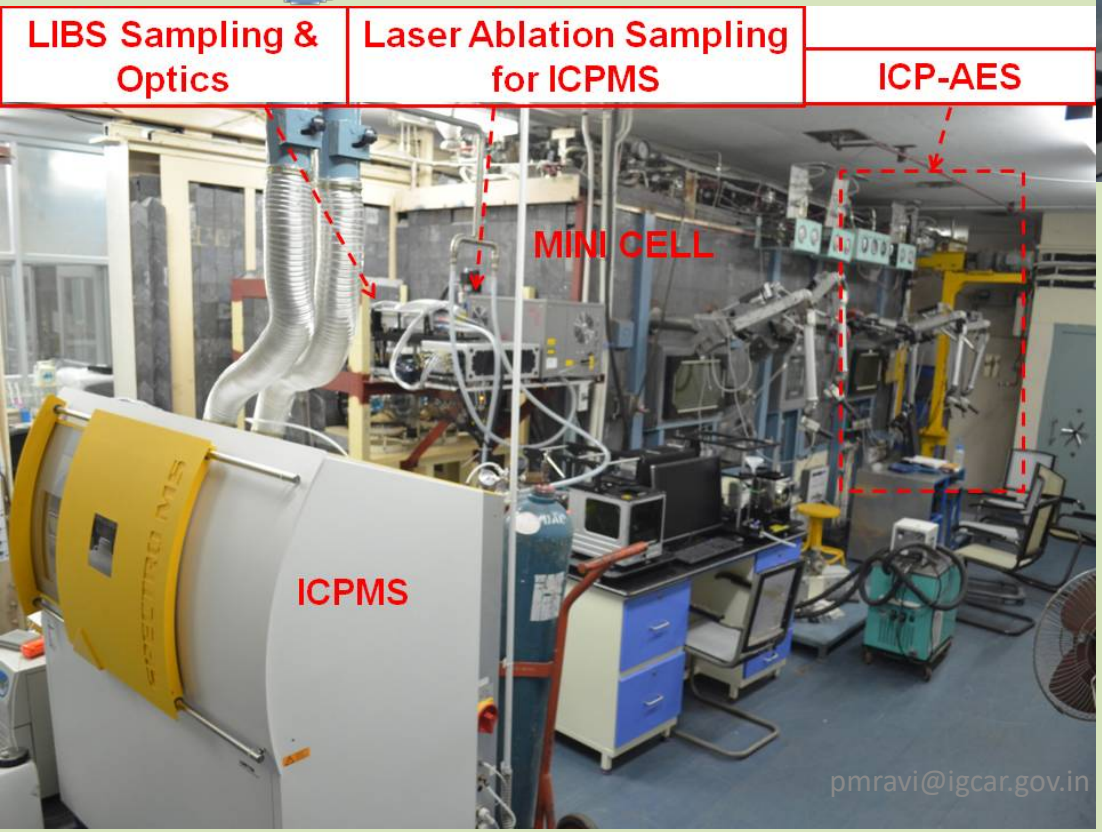
- Laser Induced Breakdown Spectroscopy (**LIBS**).
- Laser plasma light captured by CCD Detector through Spectrometer, after “plasma cooling”.
- Solid / Liquid / Gas samples
- Spatial Profiling / Surface Chemical mapping.
- Spatial isotope mapping?
- Fiber Optics allows “**Easy**” Glove Box / Hot Cell adptation.



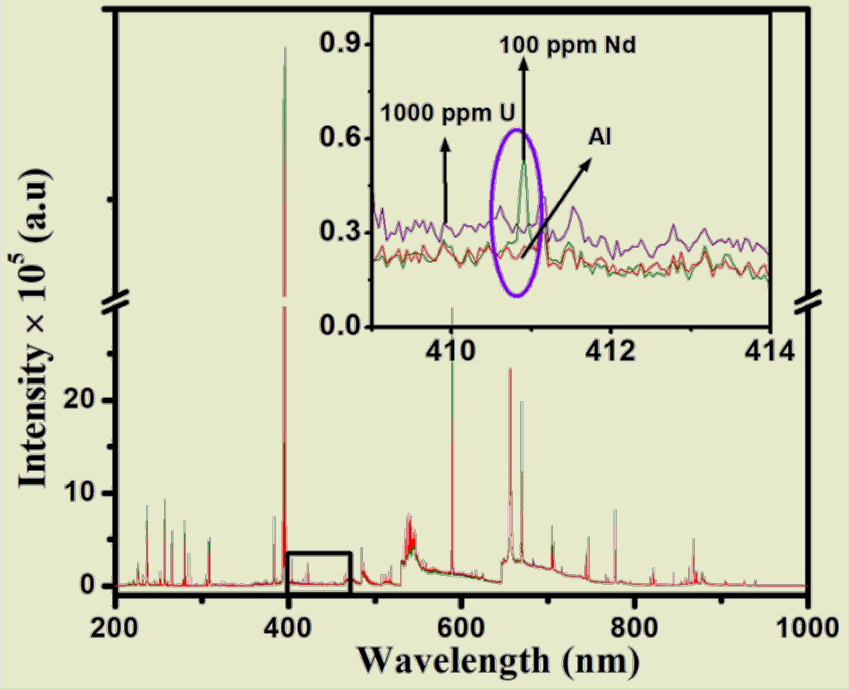
LIBS for Active Sample Analysis



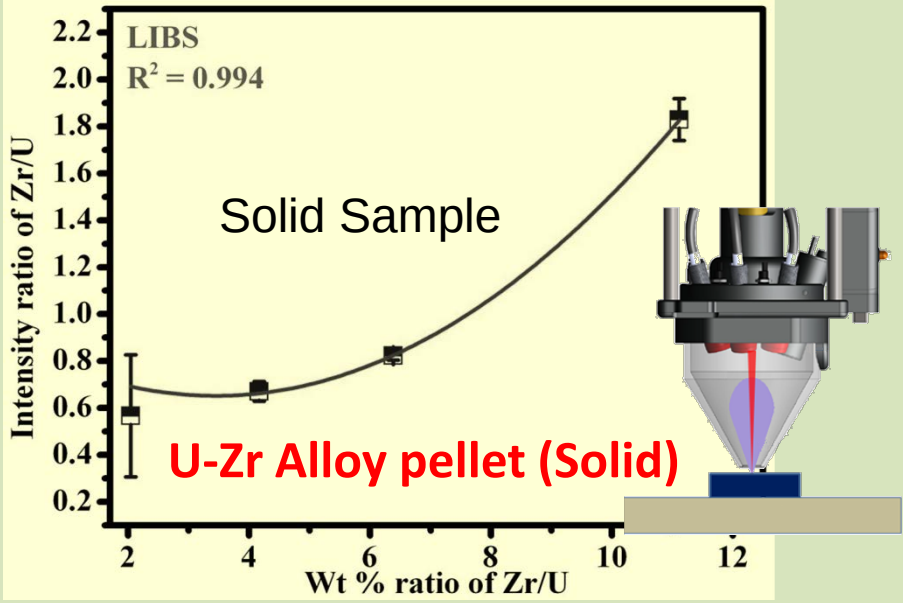


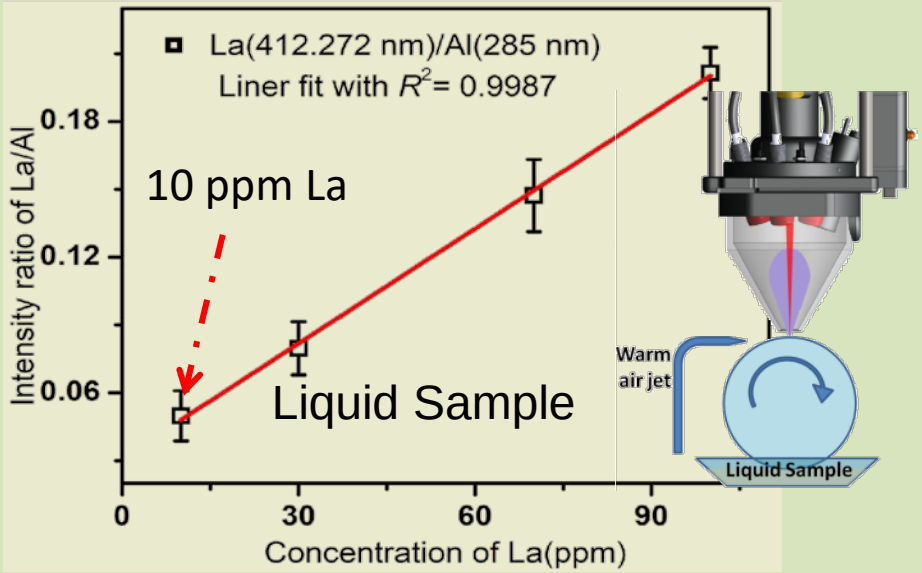


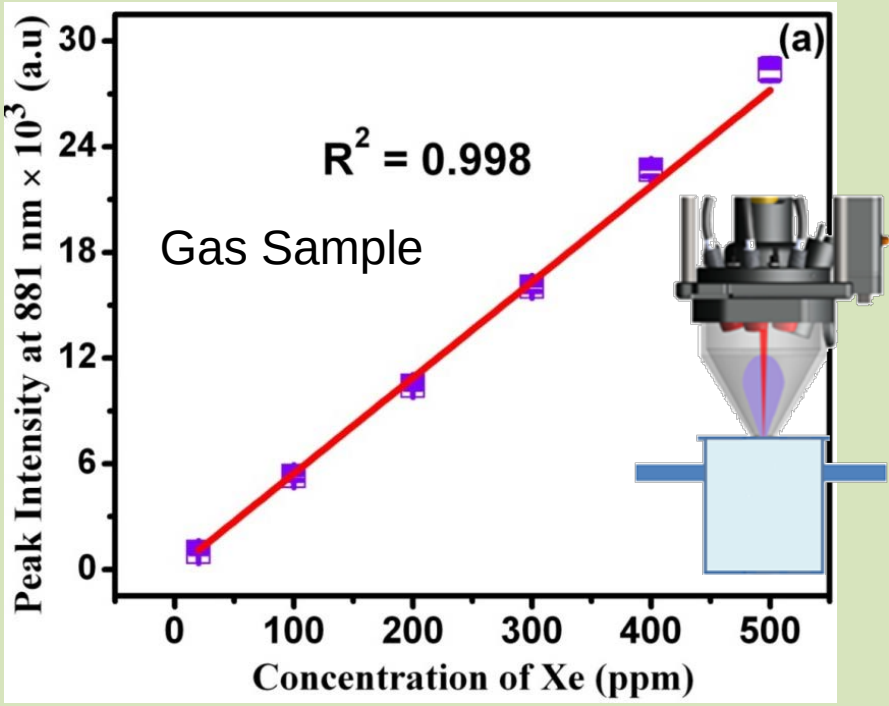
Mini Cell Facility
(In Tandem with Hot Cell Facility)
In Radio Chemistry Laboratory
(IGCAR – Kalpakkam)



LIBS Spectrum of Nd, La, Sm, U in Nitric Acid







Fission products in HLLW (Liquid)

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Fission Gas analysis (Gas)

Conclusions

Quadrupole Mass Spectrometer **QMS**
- Partial Pressure

Time of Flight Mass Spectrometer **TOF-MS**
-Isotope & elemental ratio

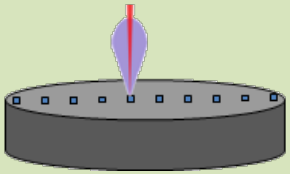
Laser Ablation Sampling

ICP – MS
-Elemental Analysis

LIBS
-Elemental Analysis

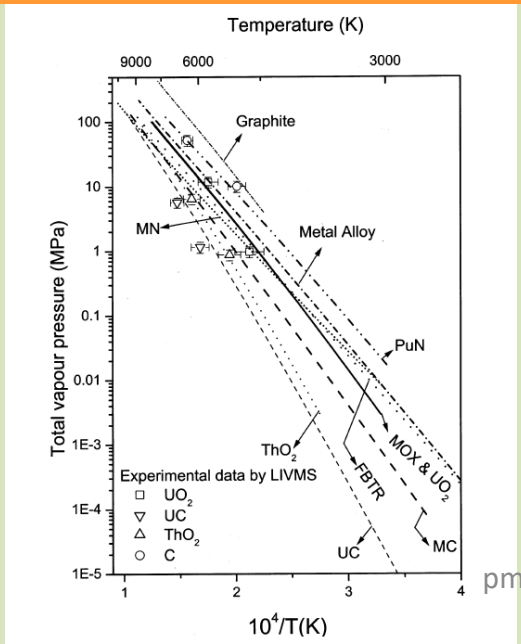
Spatial Profiling in Solid Samples

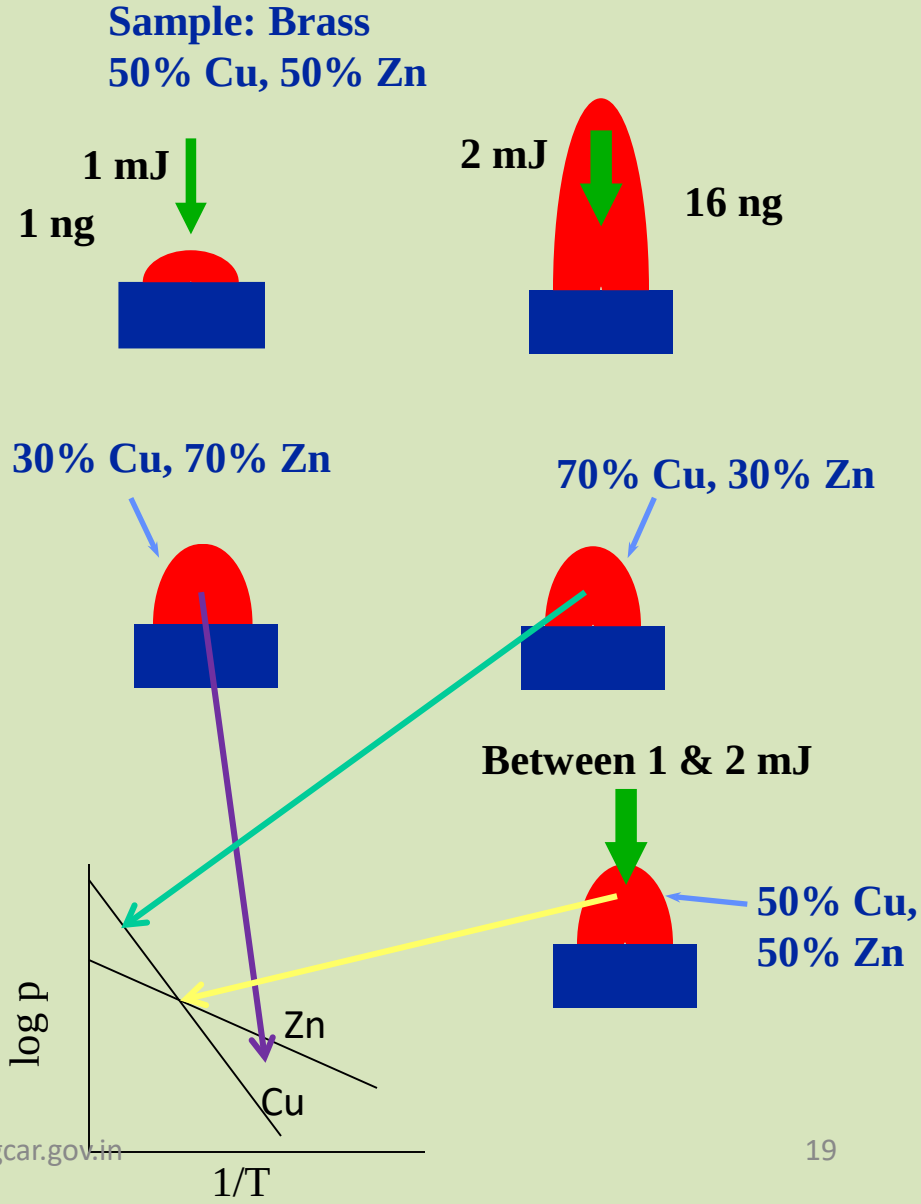
Solid, Liquid, Gas Samples



Pulsed Laser Evaporation (Laser Ablation under thermal process)

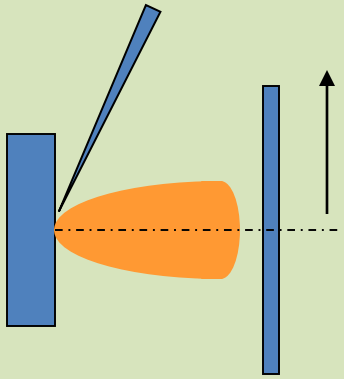
- **Quantity of ablated mass**
  - Amount of ablated mass depend on laser energy non-linearly
    - » irradiance, wavelength, pulse width
- **Composition (Chemistry) of ablated mass**
  - Composition of ablated mass is different from the solid sample
  - Composition of ablated mass and sample can be the same under particular laser conditions



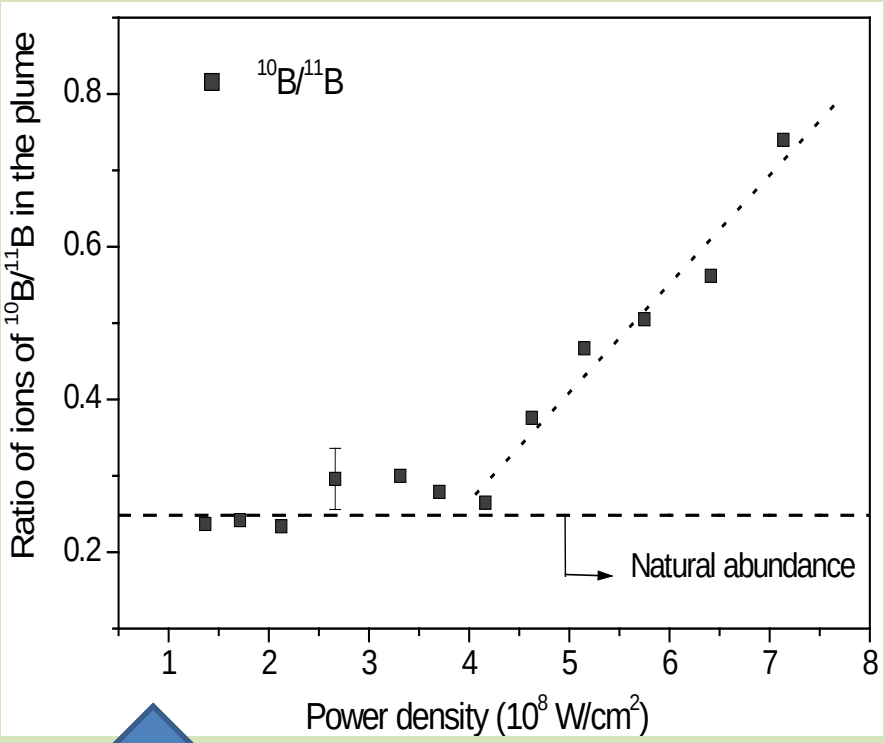


Boron isotope ratio under high laser power density

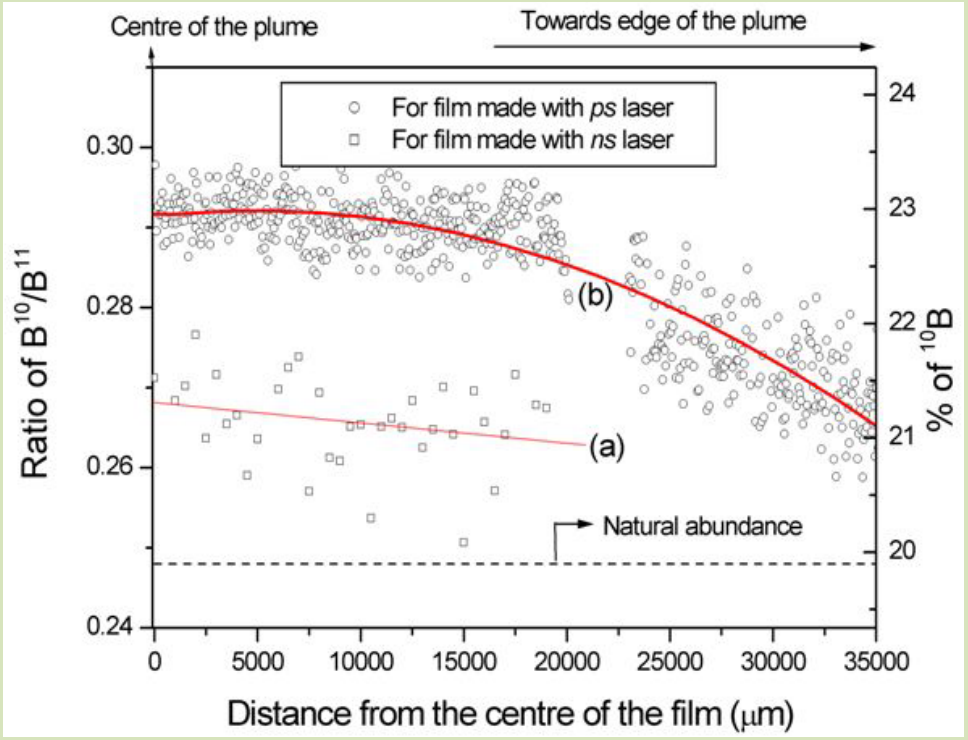
Using higher laser power > 10^8 W/cm^2



Distance from Centre of substrate



Mass Spec Studies of Laser Plume



SIMS Studies of thin film