University of California, Los Angeles

Experimental and numerical study of transient condensation of lithium and beryllium fluoride excited vapors for IFE systems

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Research Co-ordination Meeting on

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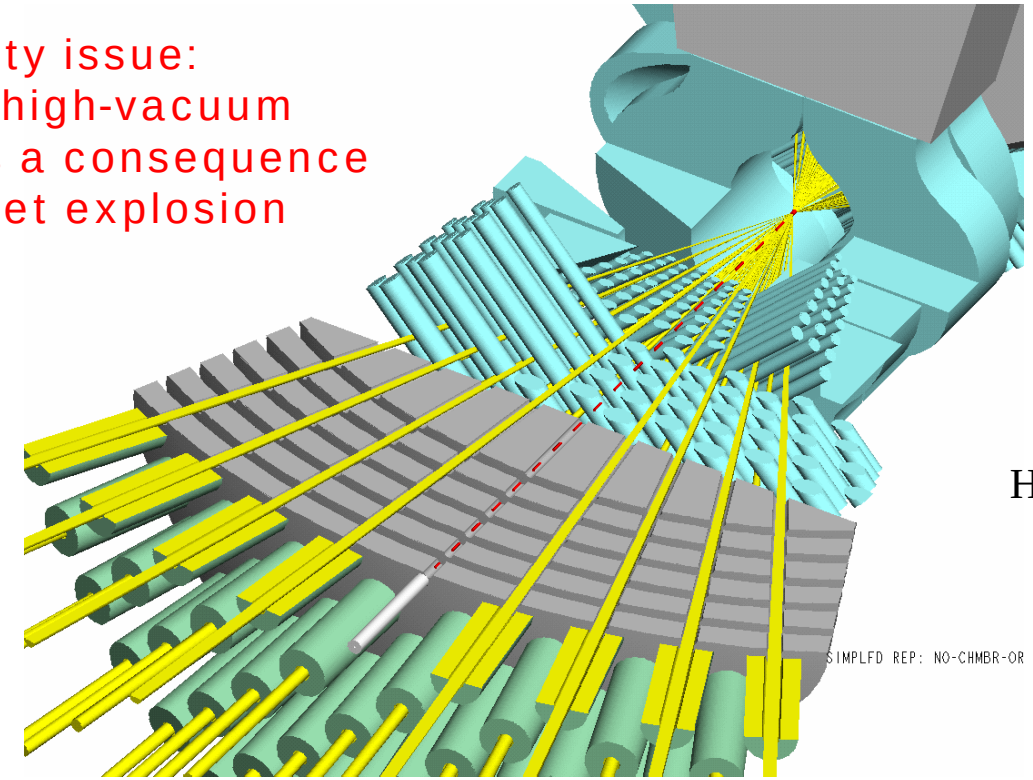
Liquid protection for IFE chambers

- Thick liquid “pocket” shields chamber structures - fluid mechanics questions replace materials questions
- Stationary jet grid protects beam lines; oscillating jets form central “pocket” and dynamically clear droplets near target
- Neutron damages and activation of flowing liquid accumulate only in the short residence period - no blanket replacement required, increases availability

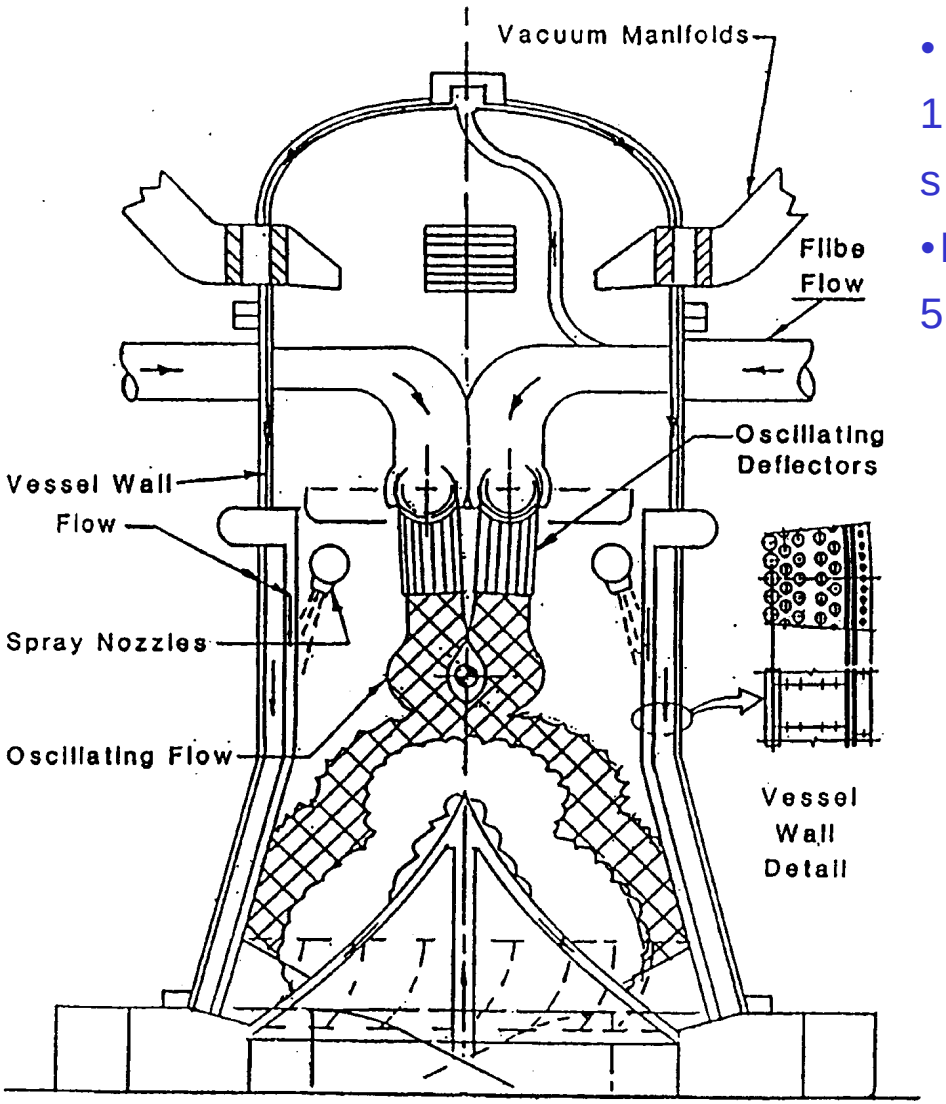
Main feasibility issue: disruption of high-vacuum conditions as a consequence of fusion target explosion

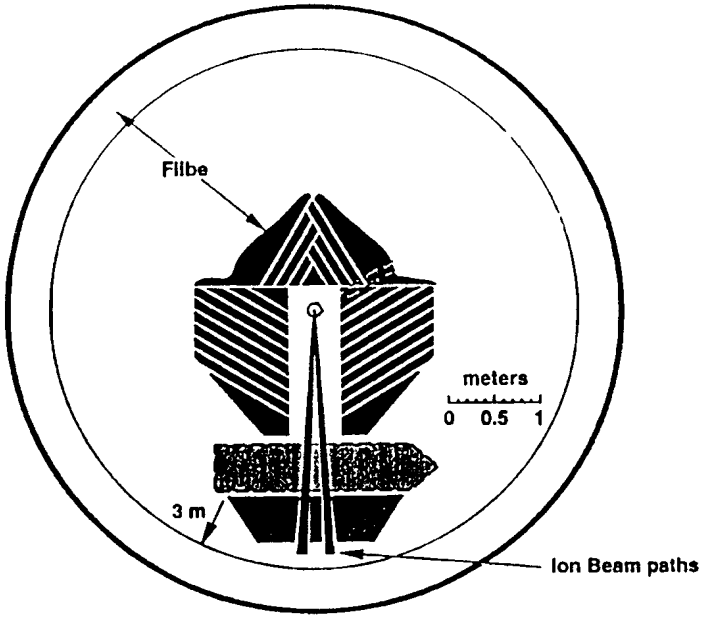


HYLIFE-II concept, LLNL drawing

HYLIFE-II design

- Chamber volume: 280 m^3
- Primary liquid loop: 150×10^3 kg/s, inlet 600 C, outlet 650 C
- Liquid pocket: 0.75 m thick, 12 m/s vertical velocity, 0.15 m spacing
- Droplet spray: 2.4×10^3 kg/s at 570 C, 2 mm diameter

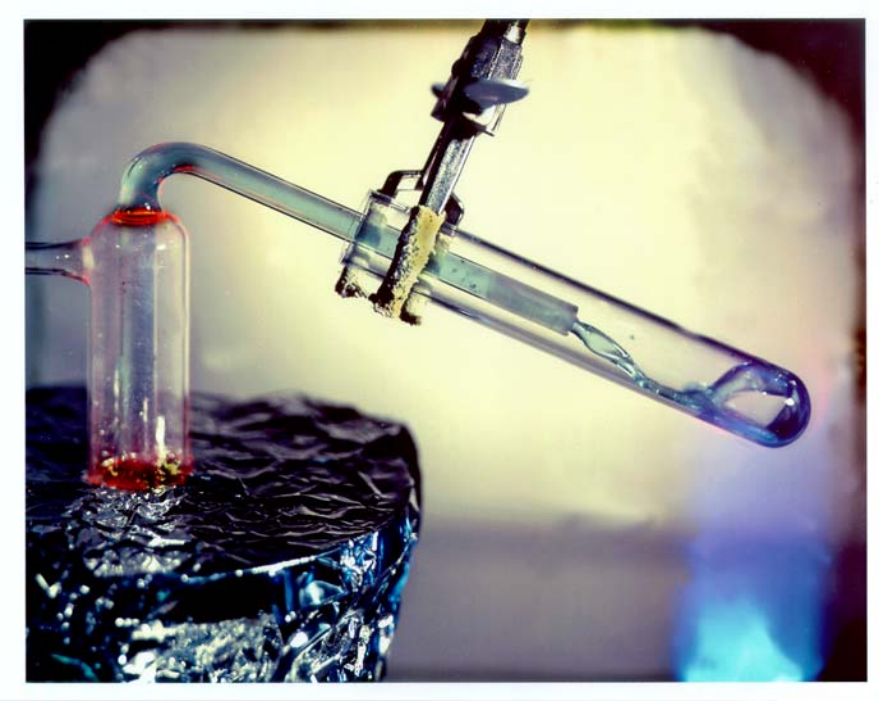




Cross Section through liquid
Top View

Flibe as liquid wall material

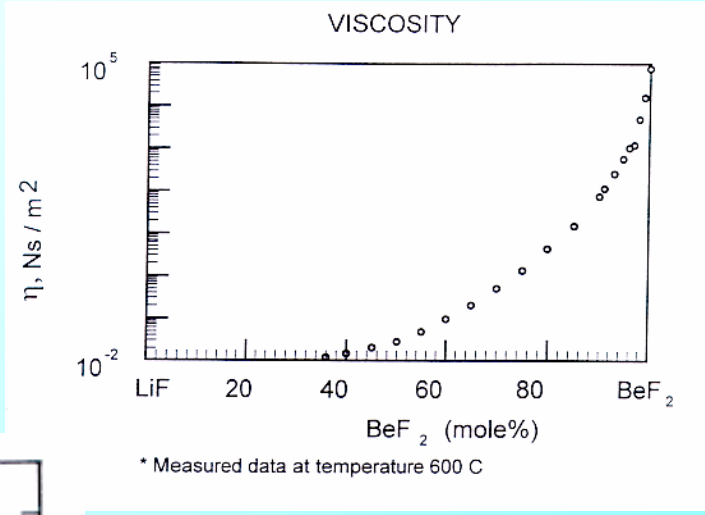
$2\ LiF + 1\ BeF_2$ (moles)

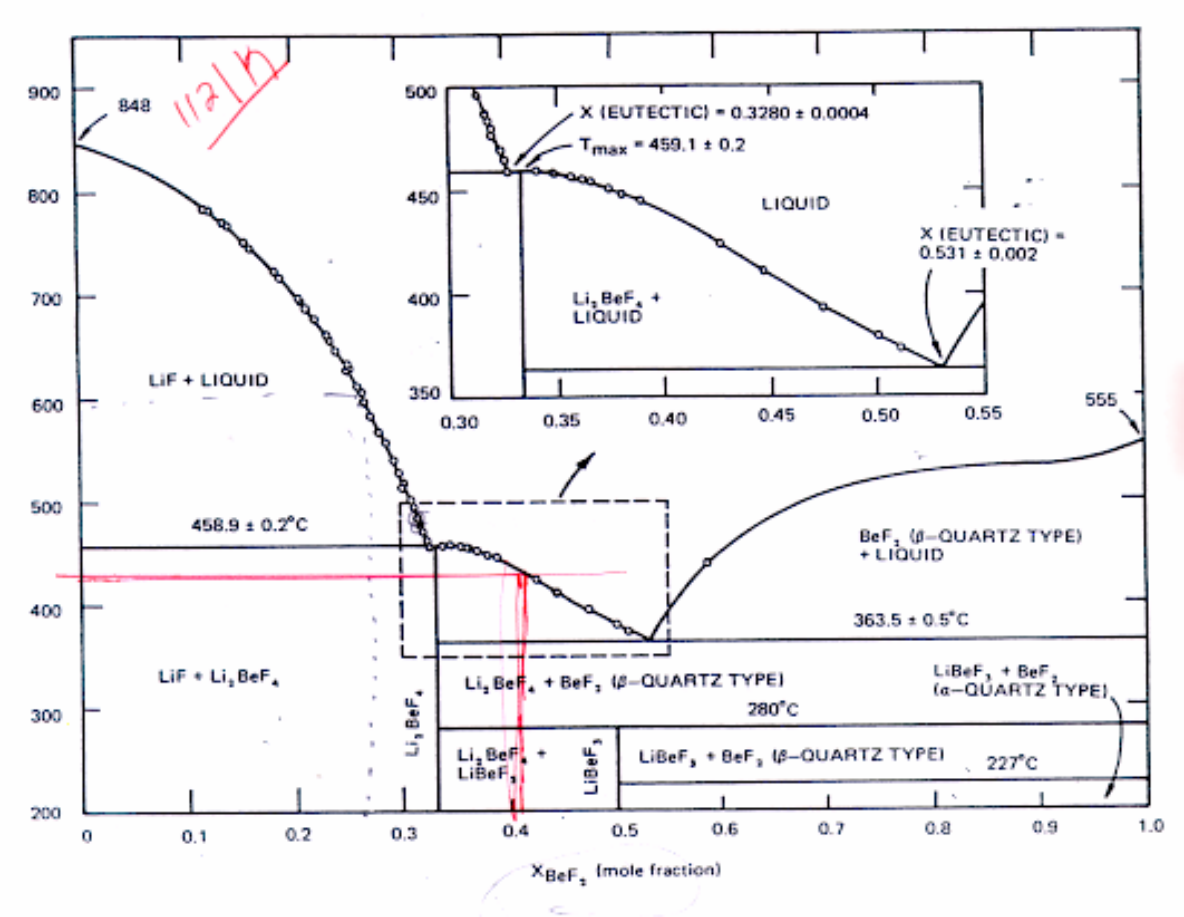


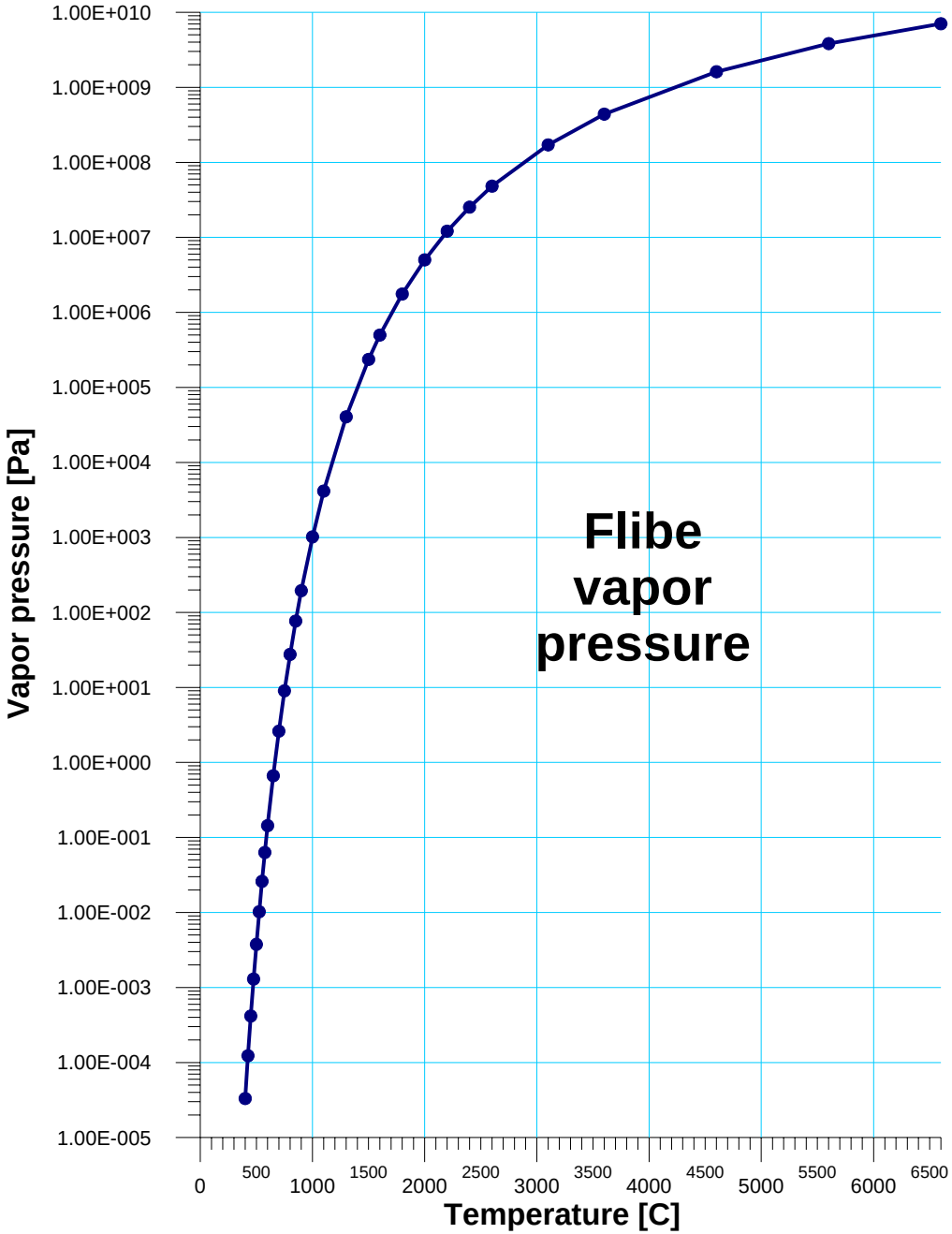
Liquid flibe flowing in a quartz pipe (courtesy of ORNL) - melting point is 460 C

Flibe crystals at UCLA are handled in a methanol bath to minimize Be particulate contamination

Flibe composition is a compromise between melting temperature (structural costs) and viscosity (pumping cost)





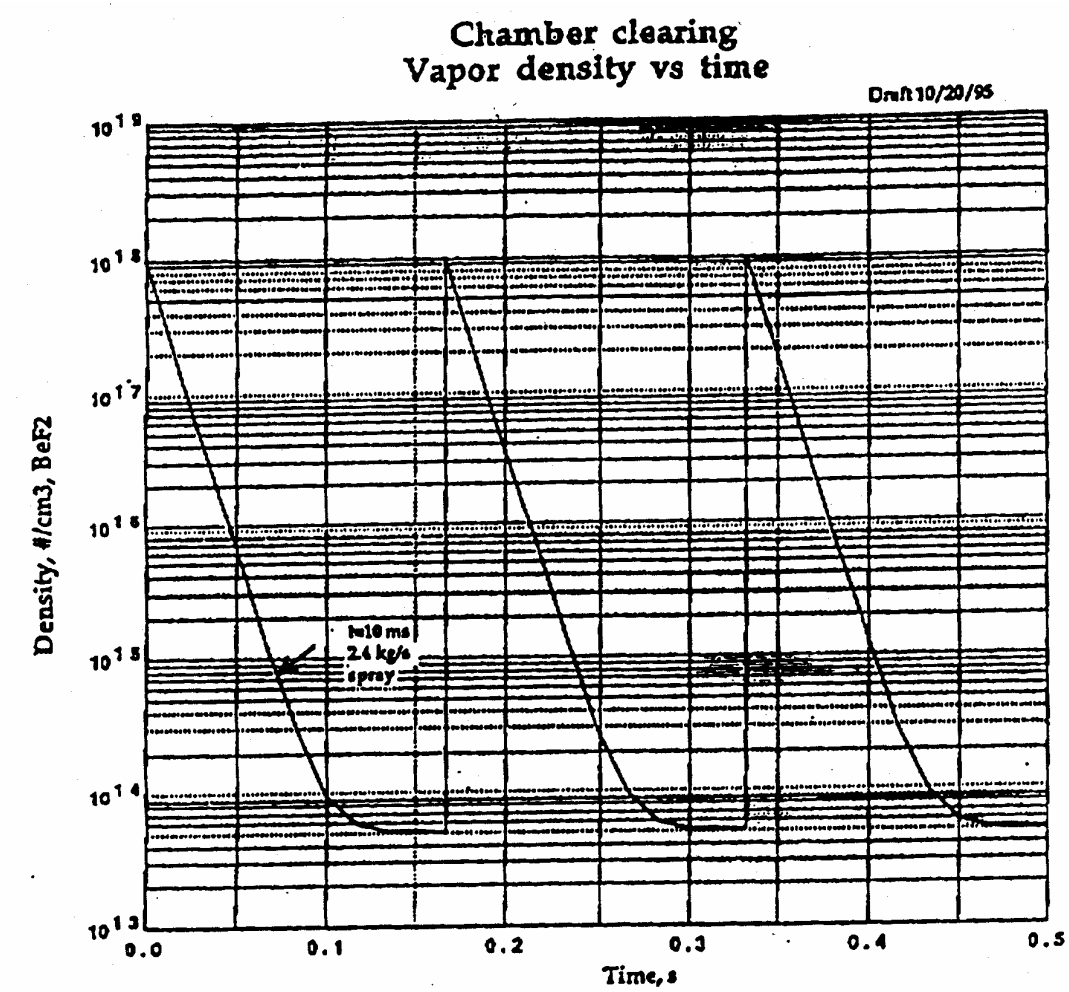


Flibe low vapor pressure is a key property for efficient driver coupling with the fusion target at high repetition rates

Reference data for flibe vapors are based on the assumption of thermodynamic equilibrium at the liquid interface and ideal gas equation

In equilibrium with the liquid at 600 C, flibe vapor pressure is about 1 mTorr, corresponding to a density of 1.2 10^{13} cm^{-3}

The objective of the experiments



R.Y. Bai and V.E. Schrock, "An Approximate method for analyzing transient condensation on spray in HYLIFE-II," Fus. Tech. 19 (1991) 732-739.

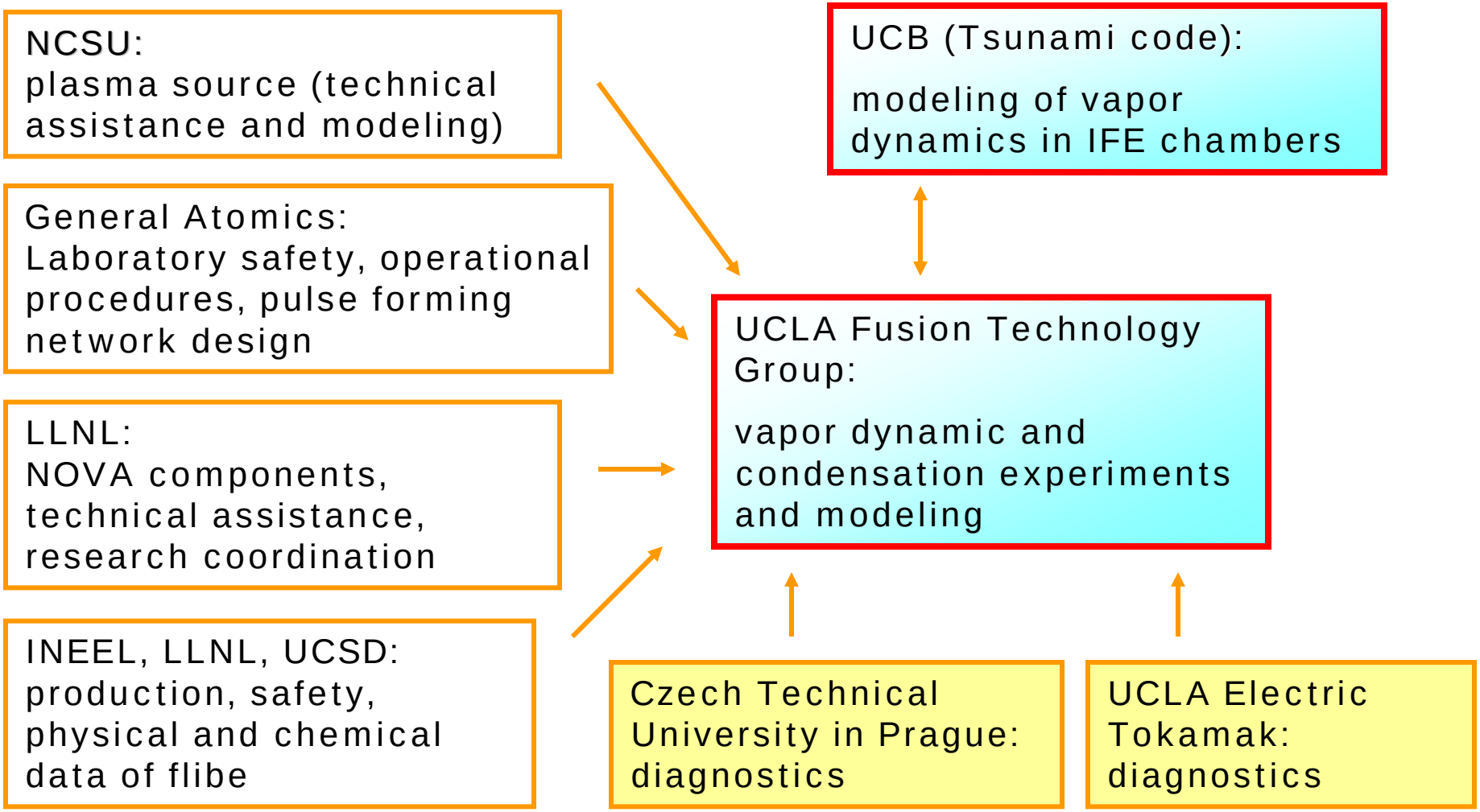
Measuring chamber clearing rates using prototypical materials

Pressure history

Investigating vapor properties at liquid / vapor interface to assess model validity and effective condensation rates

Mass spectroscopy
Emission spectroscopy

Collaboration enhances quality of research



Reproducing HYLIFE chamber conditions for condensation: experiments scaling

Assumption 1

Superheated vapor thermodynamic properties are a result of nanosecond and microsecond phenomena in the chamber, and can be assumed as initial conditions for the condensation process

Assumption 2

Condensation is a one dimensional phenomena that occurs at liquid / vapor interfaces

Scaling reduces to:

Obtain same initial conditions of superheated vapor in the experiments: particle density and temperature

For the same initial particle density, allowing the same liquid / vapor interface properties: surface area, temperature, non-condensable gases

Experiments scaling

HYLIFE chamber

Initial vapor density assuming adiabatic expansion:
Chamber volume = 280 m^3
Total estimated vaporized flibe = 14 kg
Initial particle density = 9×10^{17} #/cm^3

Initial temperature of x-ray generated vapor > than ablation mechanism - Moir's estimate for superheated vapor temperature considers pocket venting:
Initial vapor temperature = 1 eV

Since temperature range is similar, scale the initial energy coupled with the vapor, assuming all x-ray and debris absorbed by the vaporized flibe:
Initial energy density = 7.85 kJ / g

Pocket and chamber wall surface area (40 m^2) estimate not enough to ensure clearing rates > 6 Hz:
Liquid surface with droplet enhancement = 1100 m^2
Surface per unit mass = 785 cm^2 / g

Experiments

In LiF experiments total ablated mass in the source was measured:
LiF chamber volume = 5000 cm^3
Flibe chamber volume = 450 cm^3
LiF ablated mass = 0.4 g
Flibe vaporized mass = 0.1 g (?)

From NCSU measurement and modeling of electro-thermal superheated vapor:
Initial vapor temperature = 1 eV

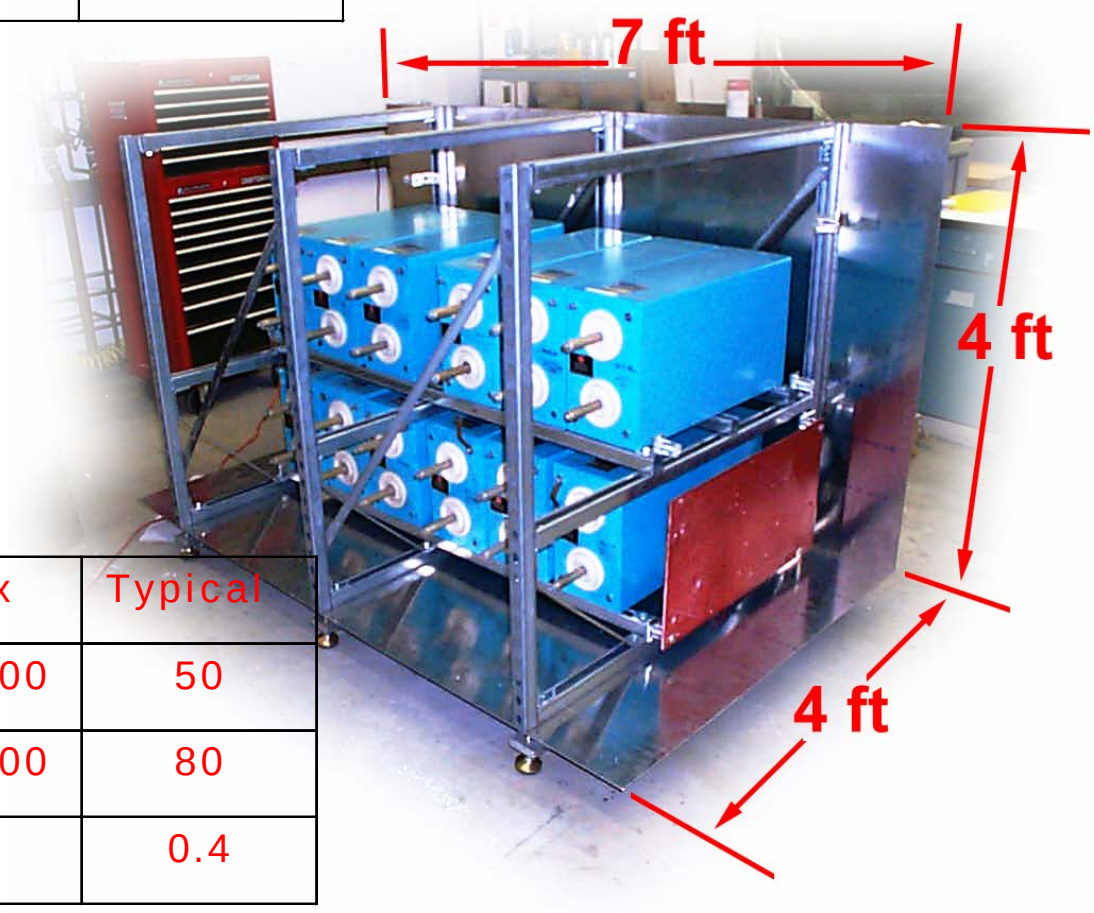
Considering a typical 4 kV discharge, total energy stored is 3.7 kJ, energy coupled with vapor is 3.0 kJ:
Initial energy density = 7.5 kJ / g

LiF ch. wall surface area = 1720 cm^3
Surface per unit mass = 4300 cm^2 / g
Flibe ch. wall surface area = 1720 cm^3
Surface per unit mass = 5100 cm^2 / g

Energy storage system

Facility characteristics

		Max	Typical
Voltage	kV	22	6
Capacitance	µF	640	320
Stored energy	kJ	154	6



Facility performance

		Max	Typical
Current peak	KA	~ 300	50
Discharge period	µs	~ 200	80
Vapor generated	g		0.4

LiF experiments: the electro-thermal source

Characterization of superheated vapor generation shows measured parameters similar to SIRENS

Typical vapor parameters in the source :

$n = 10^{19} - 10^{20}$ # / cm^3
T = 1-3 eV

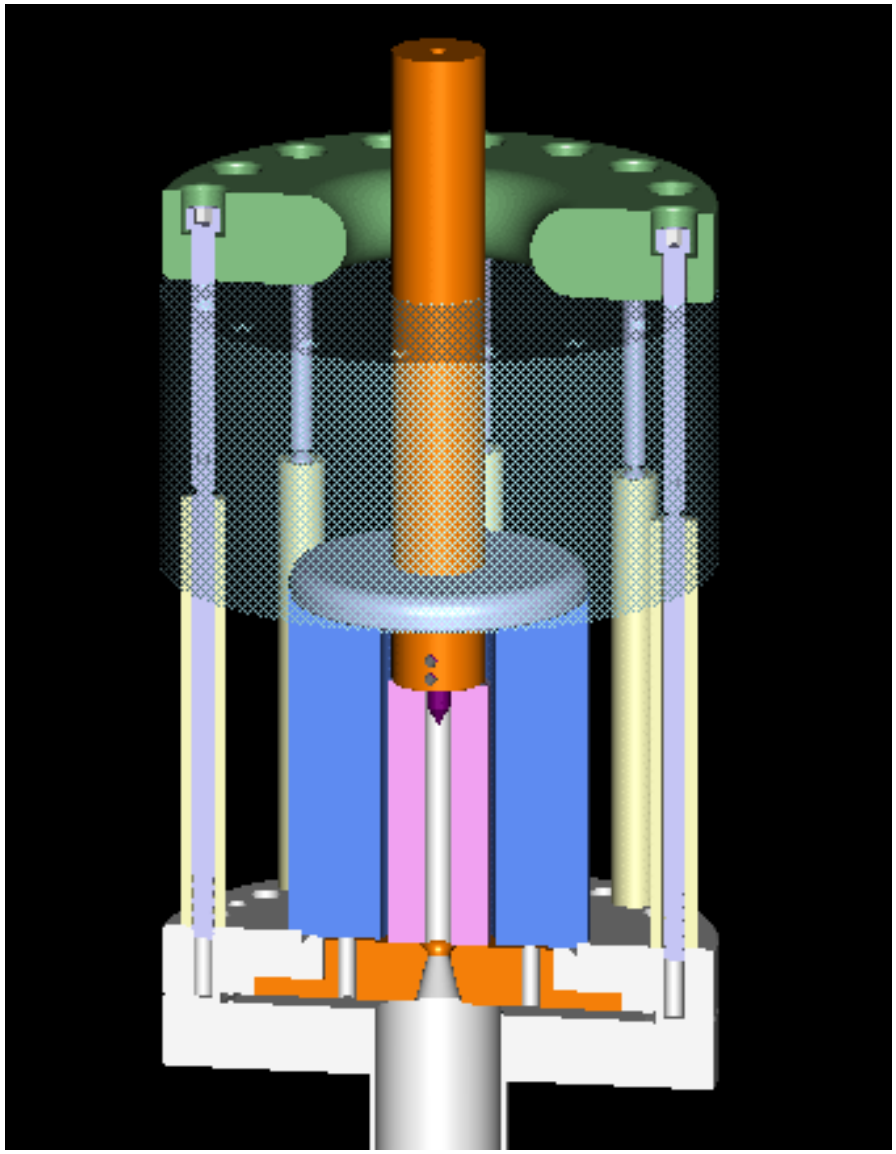
Close Ignitron switch

Cu triggering wire vaporizes (10-100 nanos) forming initial plasma column

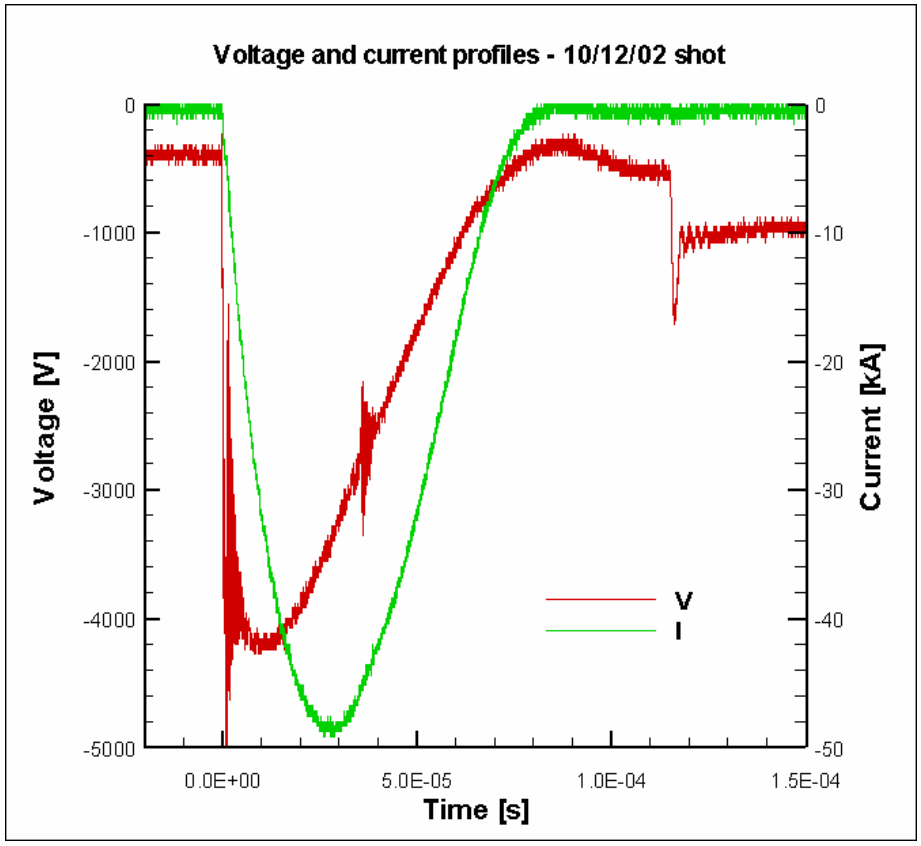
Energy stored in cap banks maintains plasma at 1-3 eV for 100 micros

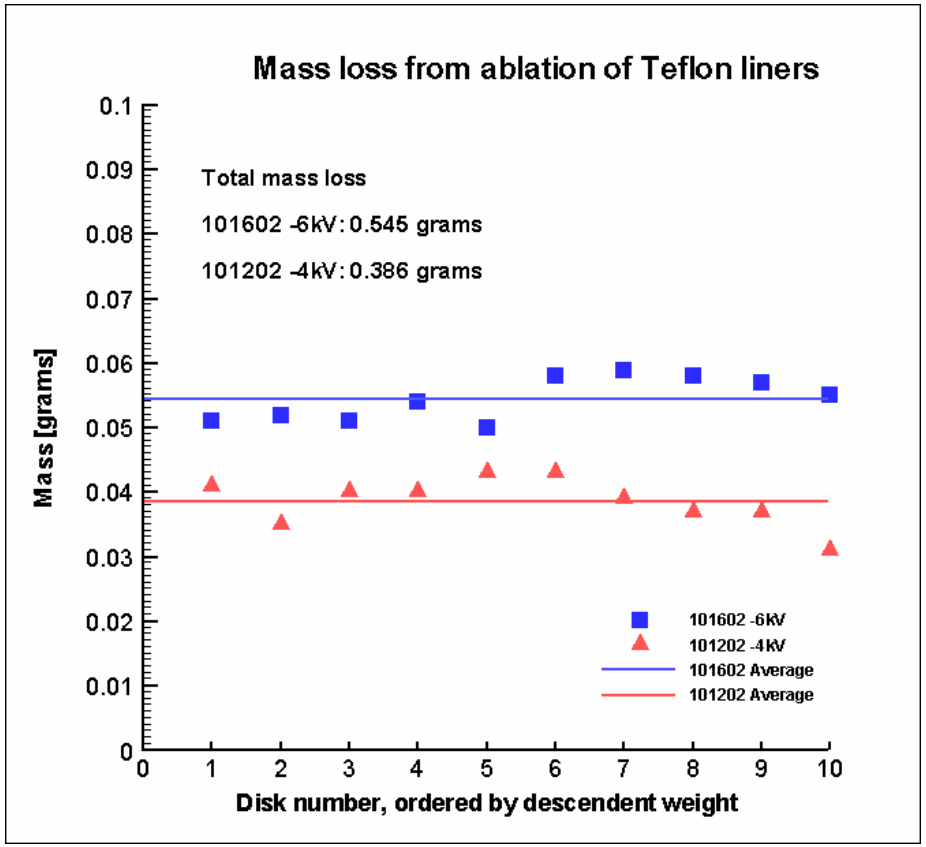
Injected electrical power radiated to surface, ablates material of interest

Pressure gradient drives injection, ablation balances axial mass loss



Measured discharge parameters





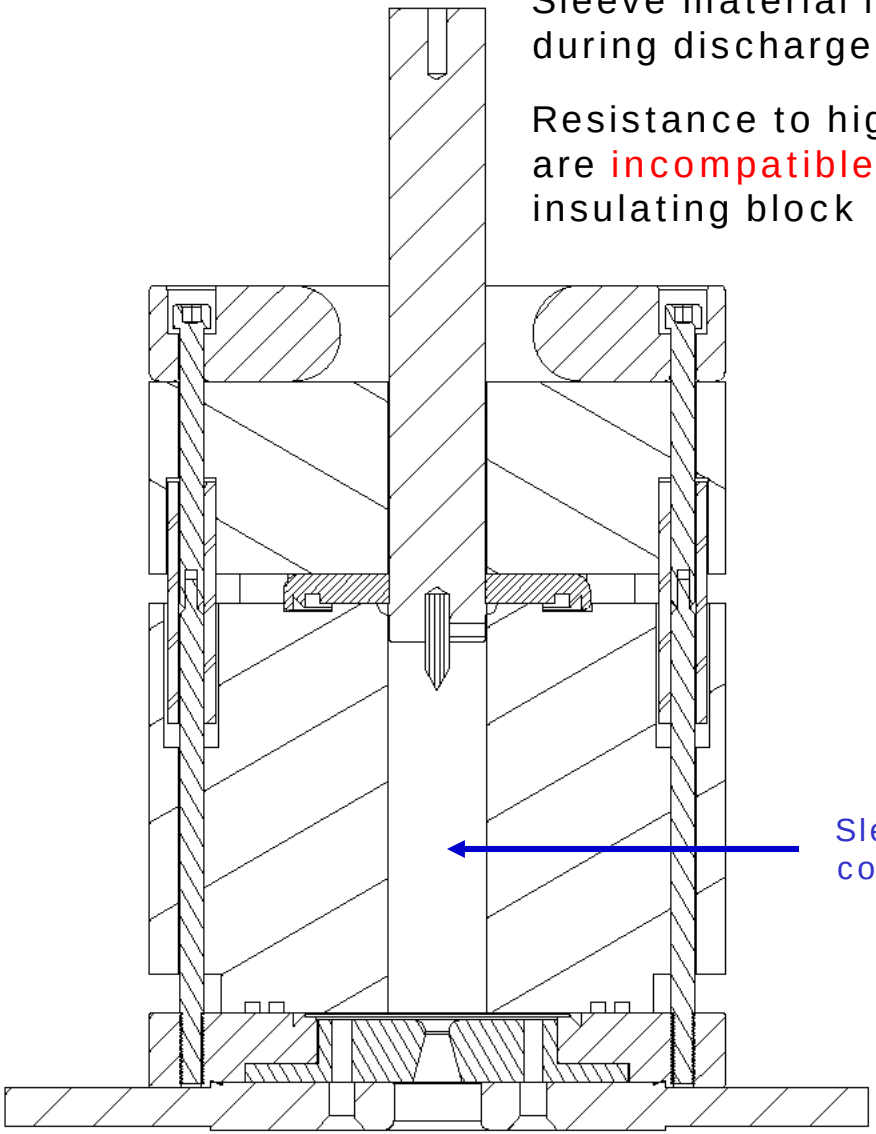
Measured electrical parameters of a typical discharge for LiF experiments

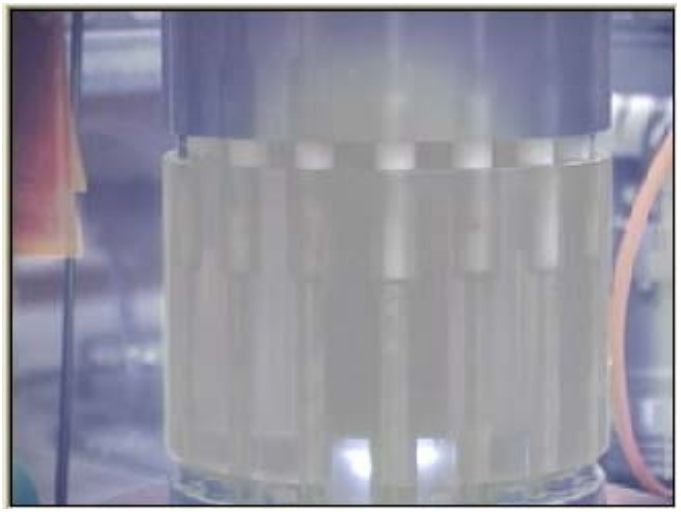
Measured mass loss from sleeve material - CF_2 results are better characterized then LiF

Limitations of the electro-thermal source

Sleeve material is responsible for plasma confinement during discharge, necessary for injection into the chamber

Resistance to high pressure peak and electrical insulation are incompatible with high temperature materials for main insulating block

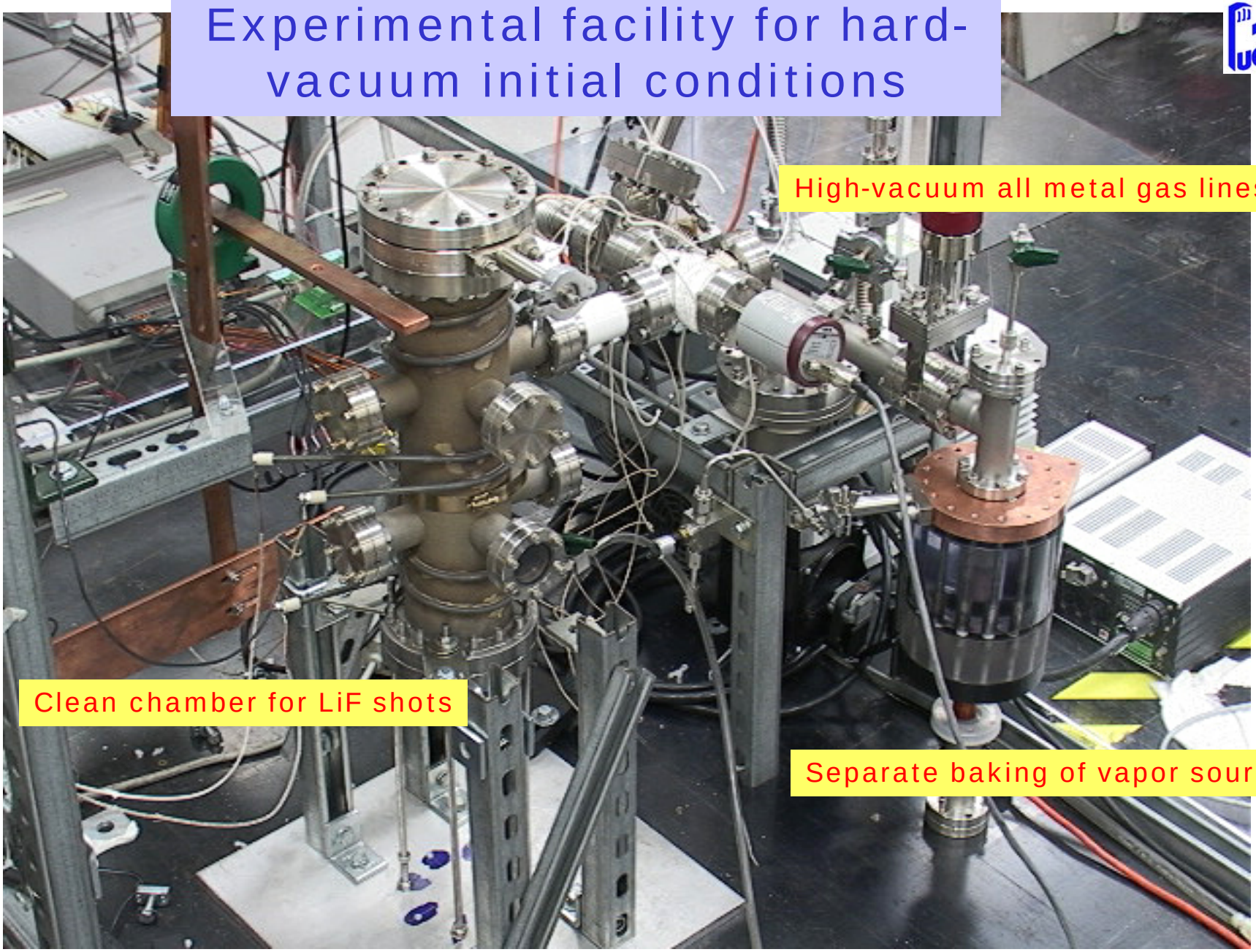


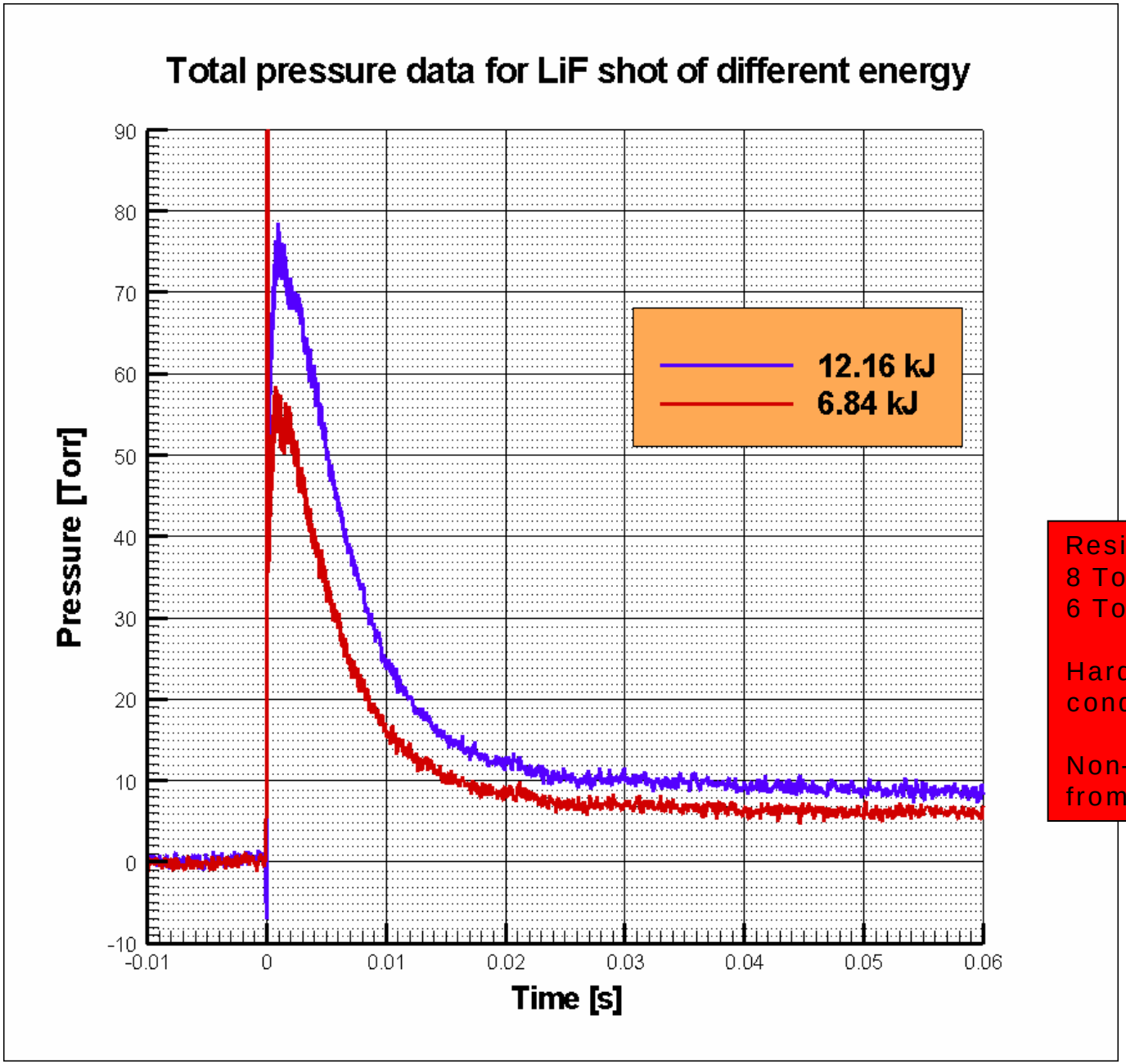


Sleeve (LiF) configuration

Fragility of LiF crystals compromises its functionality - main insulator is also ablated

Experimental facility for hard-vacuum initial conditions





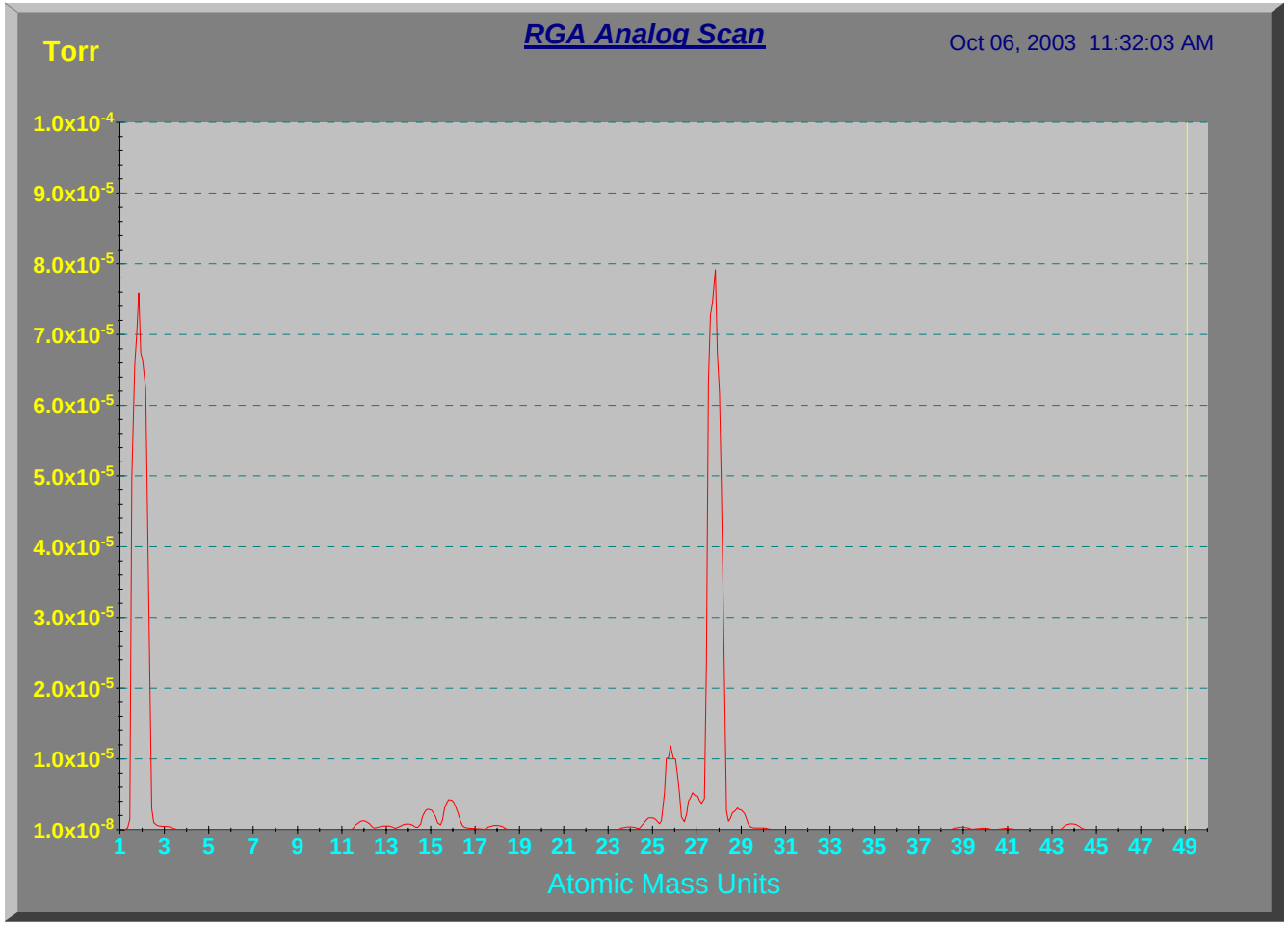
Results: pressure history

Residual gas pressure:
8 Torr
6 Torr

Hard-vacuum initial conditions are ensured

Non-condensable gases from insulator block

Mass spectroscopy for residual gases composition

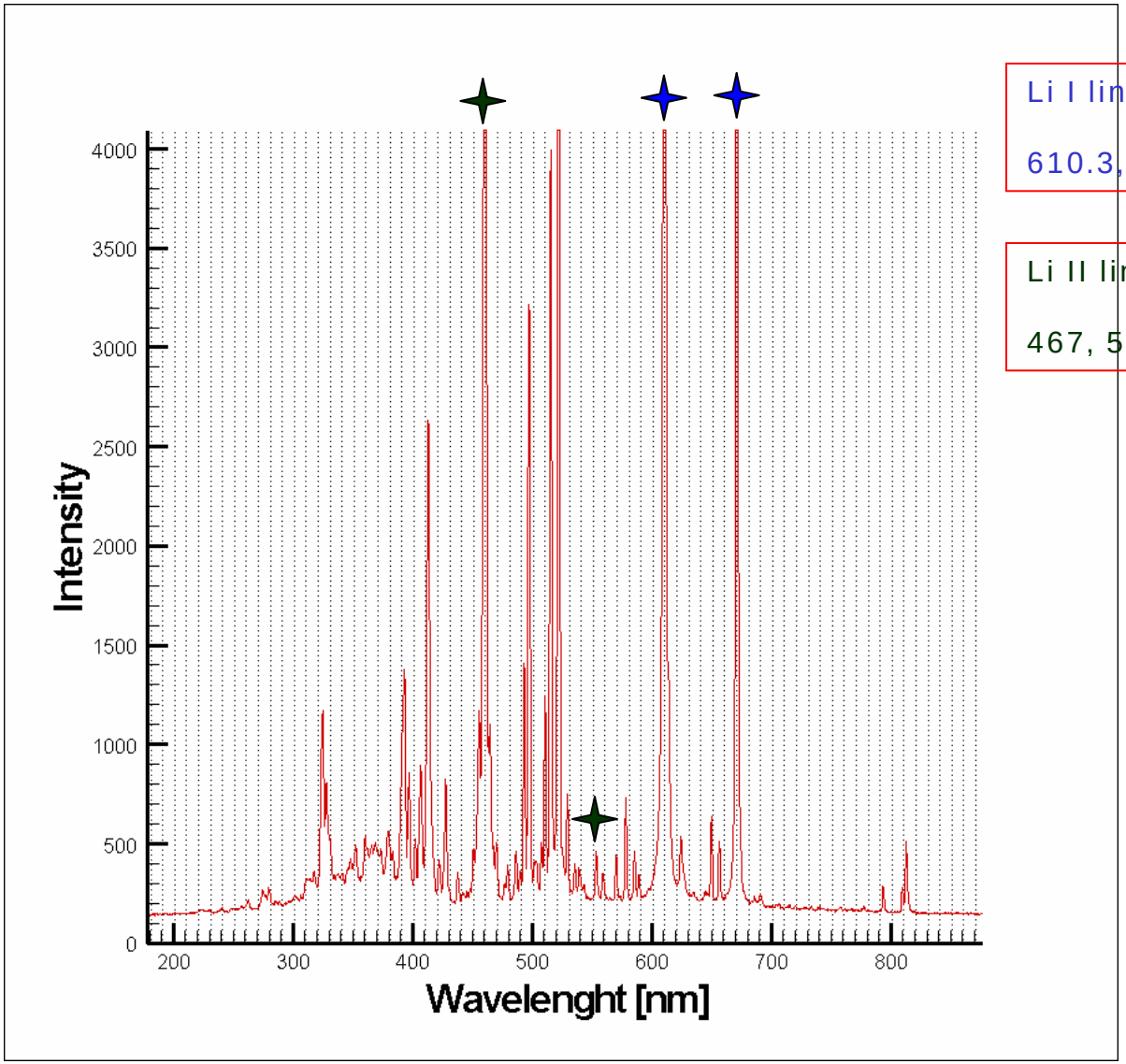


Non-condensable gases from the ablation of polycarbonate (Lexan) insulator:

H_2
CO
CO_2

300 mTorr of residual LiF

Additional diagnostics: emission spectroscopy



Li I lines:

610.3, 670.8 nm

Li II lines:

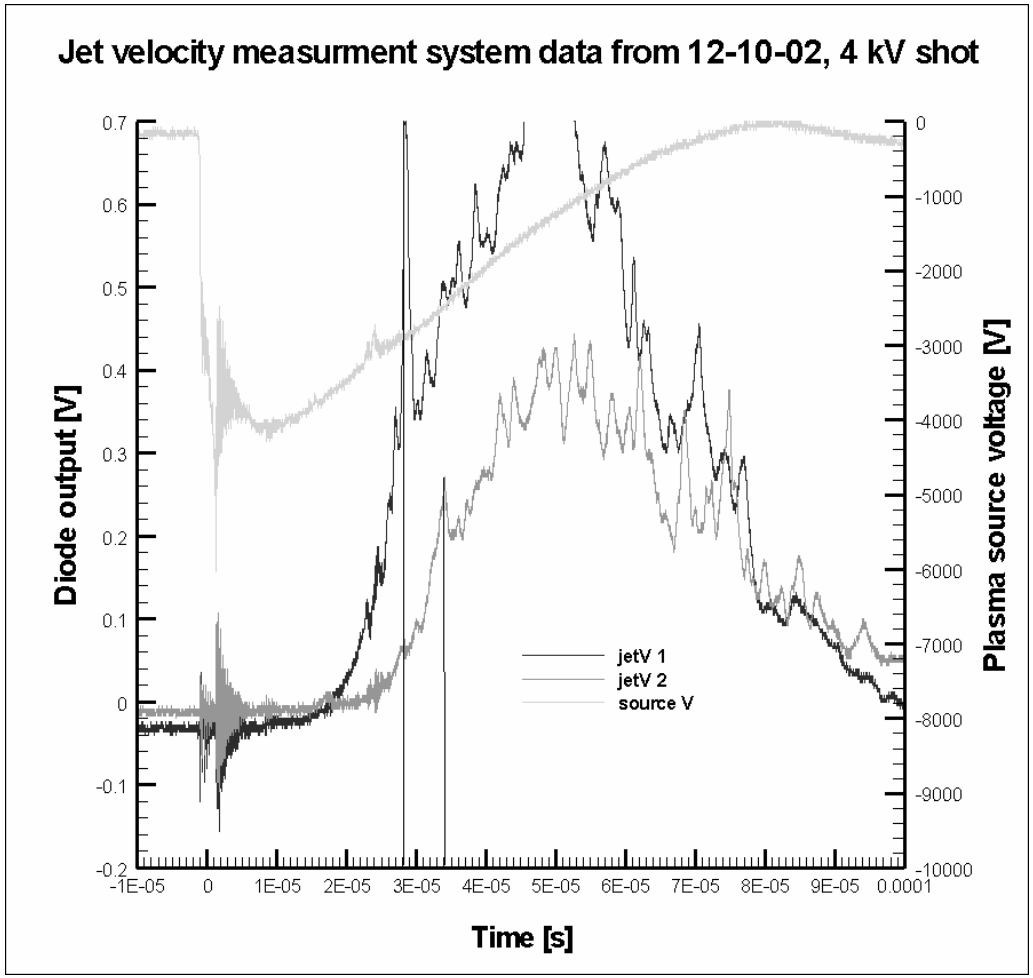
467, 548.3 nm

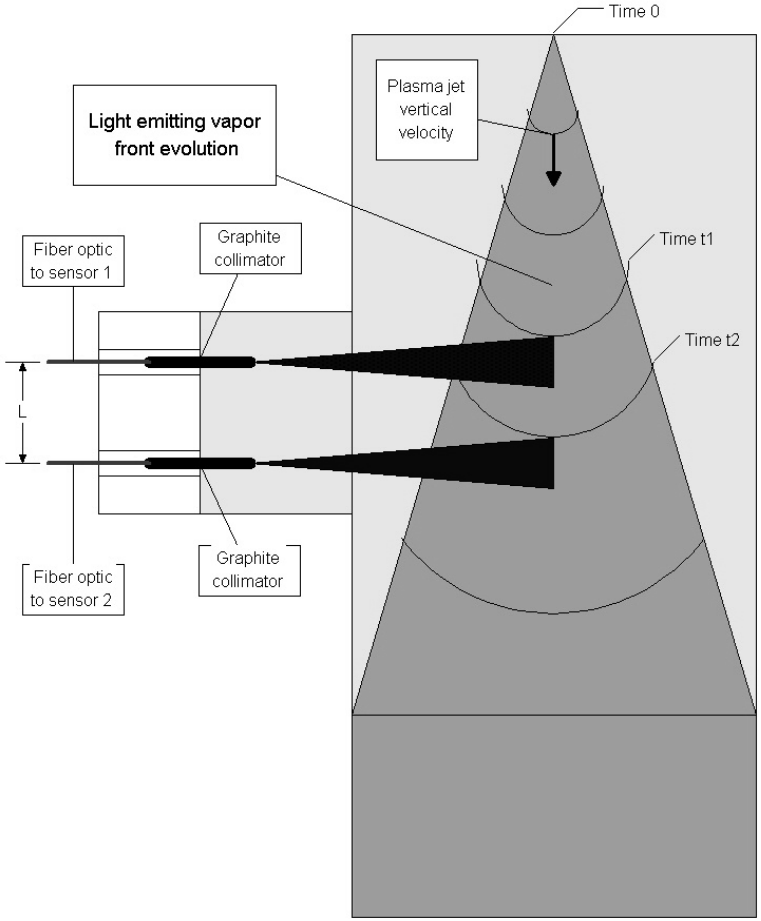
Objective:

- measure at different times the ratio of line intensity of Li I and Li II transitions

- steady-state emission calibration with langmuir probe will associate line ratio with thermodynamic vapor parameters (pressure and density)

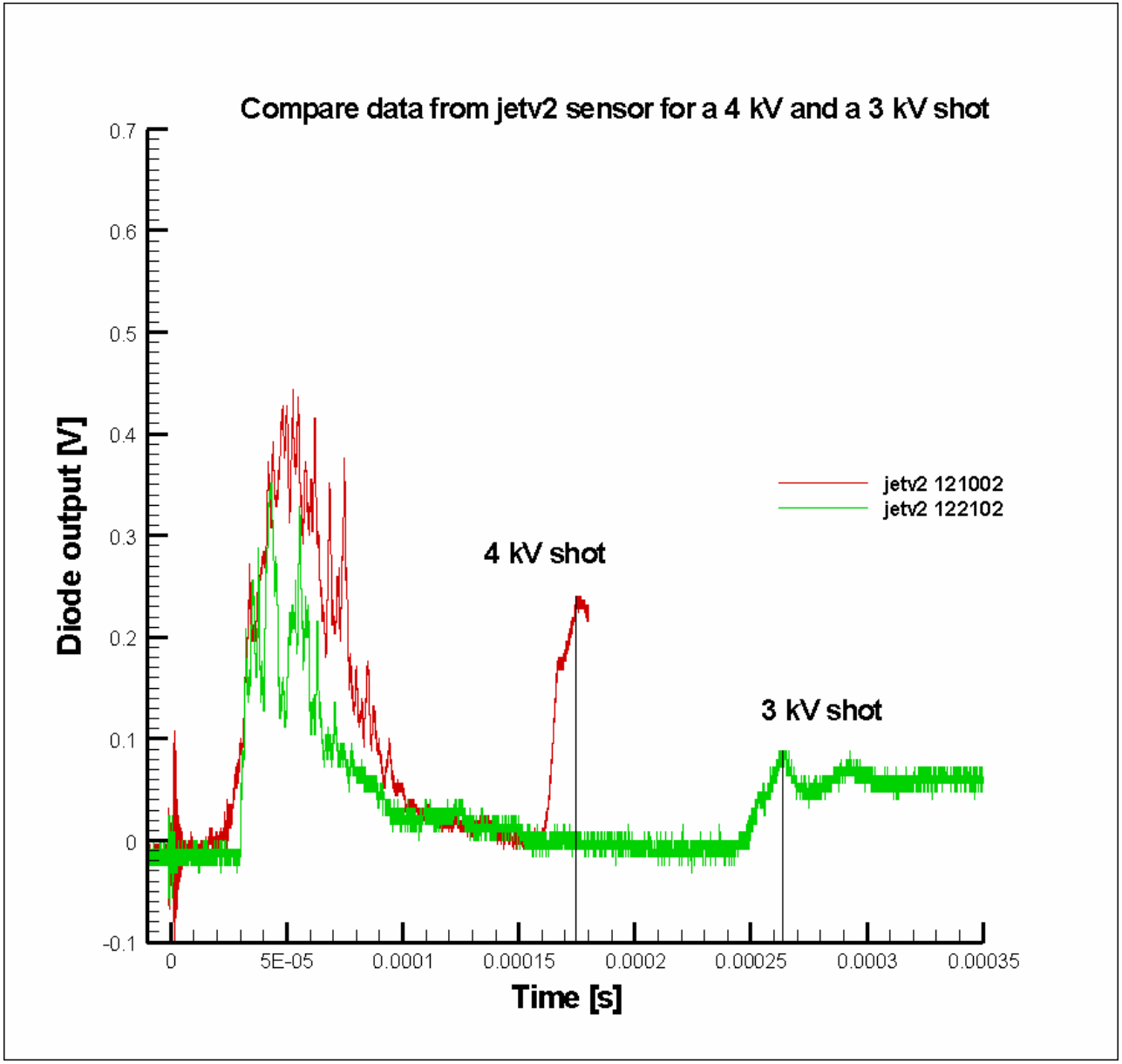
Jet velocity optical measurement system





Diode axis separation:
7.62 cm
Peak time delay:
6 microseconds
Estimated initial vapor velocity:
12700 m/s

Jet velocity optical measurement system



Sensor closer to center of chamber sees a second peak in the light, about 2ms after triggering

Peak due to first reflection of the vapor jet at the chamber bottom

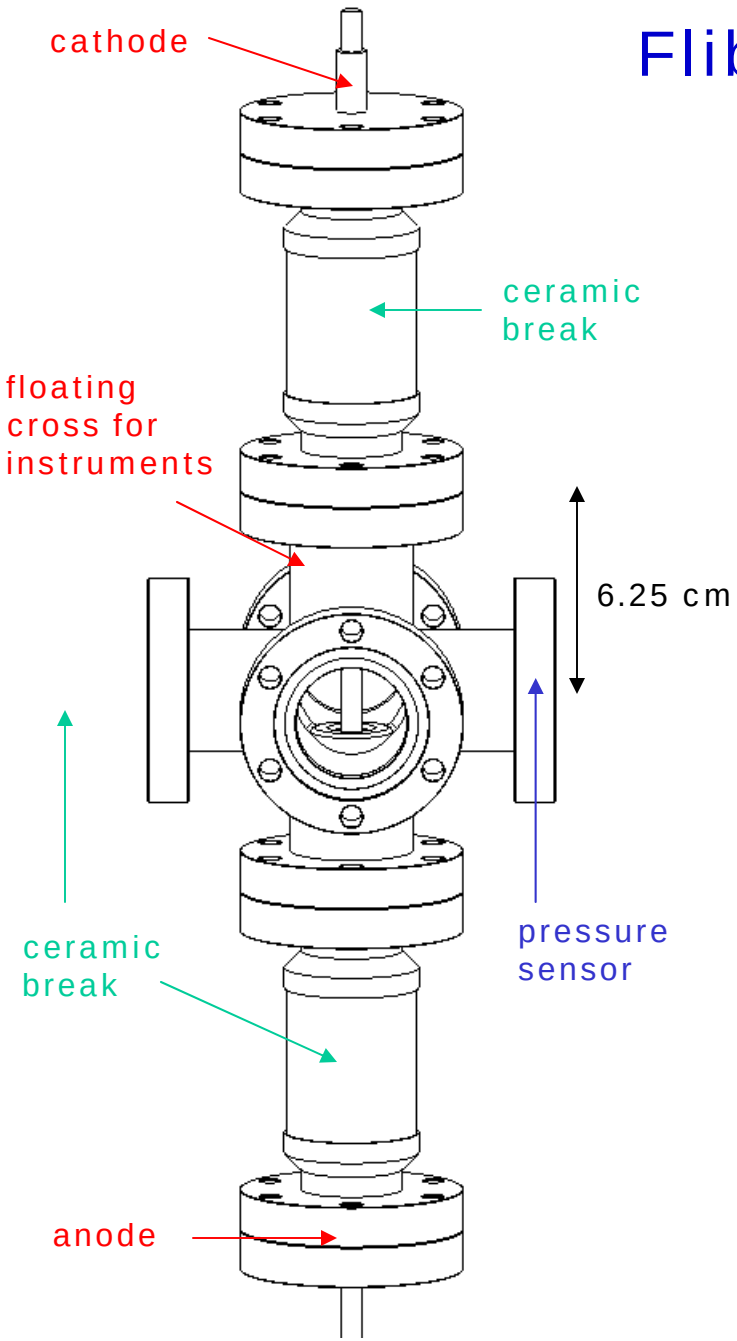
Vapor cools and expands in the chamber, emitting front does not reach the upper sensor

Estimated average velocity of vapor in the chamber after first reflection:
320 m/s - 4 kV
210 m/s - 3 kV

What we have learned from LiF experiments

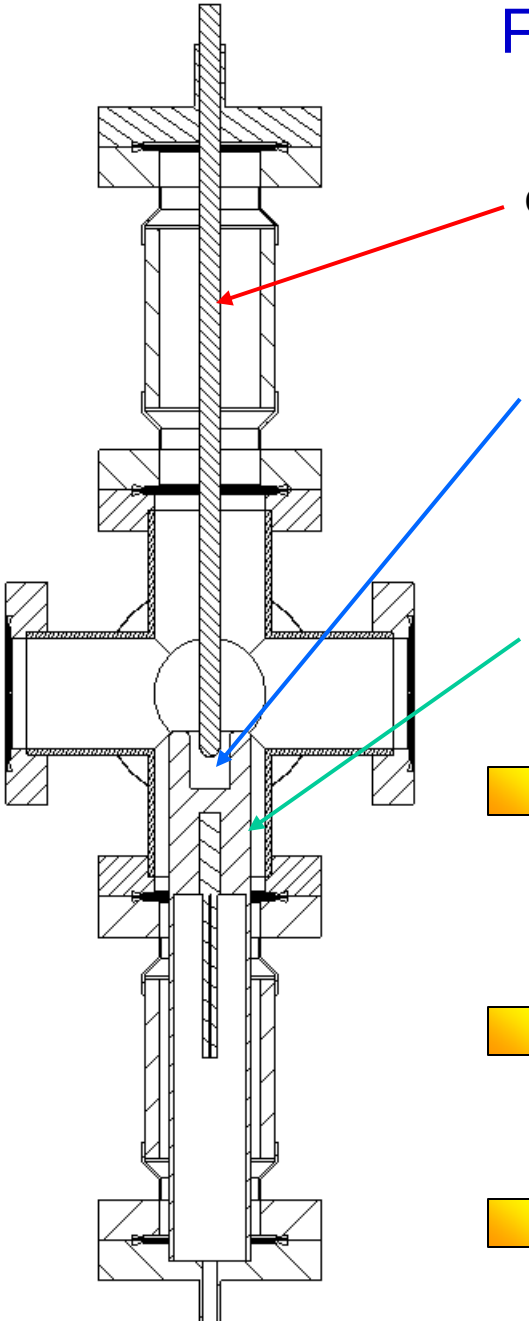
- Transient condensation of superheated LiF vapors is completed in about 20 milliseconds, compatible with IFE clearing periods
- The presence of a high amount of non condensable gases inhibits the complete condensation of LiF vapors
- The amount of non condensable however does not influence appreciably the condensation rate
- The diagnostic system is reliable and experiments can be safely carried out up to 12 kJ
- The ablating vapor source will always be associated with non condensable impurities when used with brittle solid sleeves such as molten salts that are not capable of confining the arc

Flibe experimental set-up



- Excited vapor is generated inside the condensation volume - no confining materials for injection
- Insulation is achieved with high-vacuum ceramic breaks - no plastic
- Pressure sensor is close to flibe vapor source
- Vapor is generated by vaporization of a liquid flibe surface - IFE conditions
- The conductivity of liquid flibe is the trigger for the initiation of the arc - no triggering wires

Flibe experimental set-up



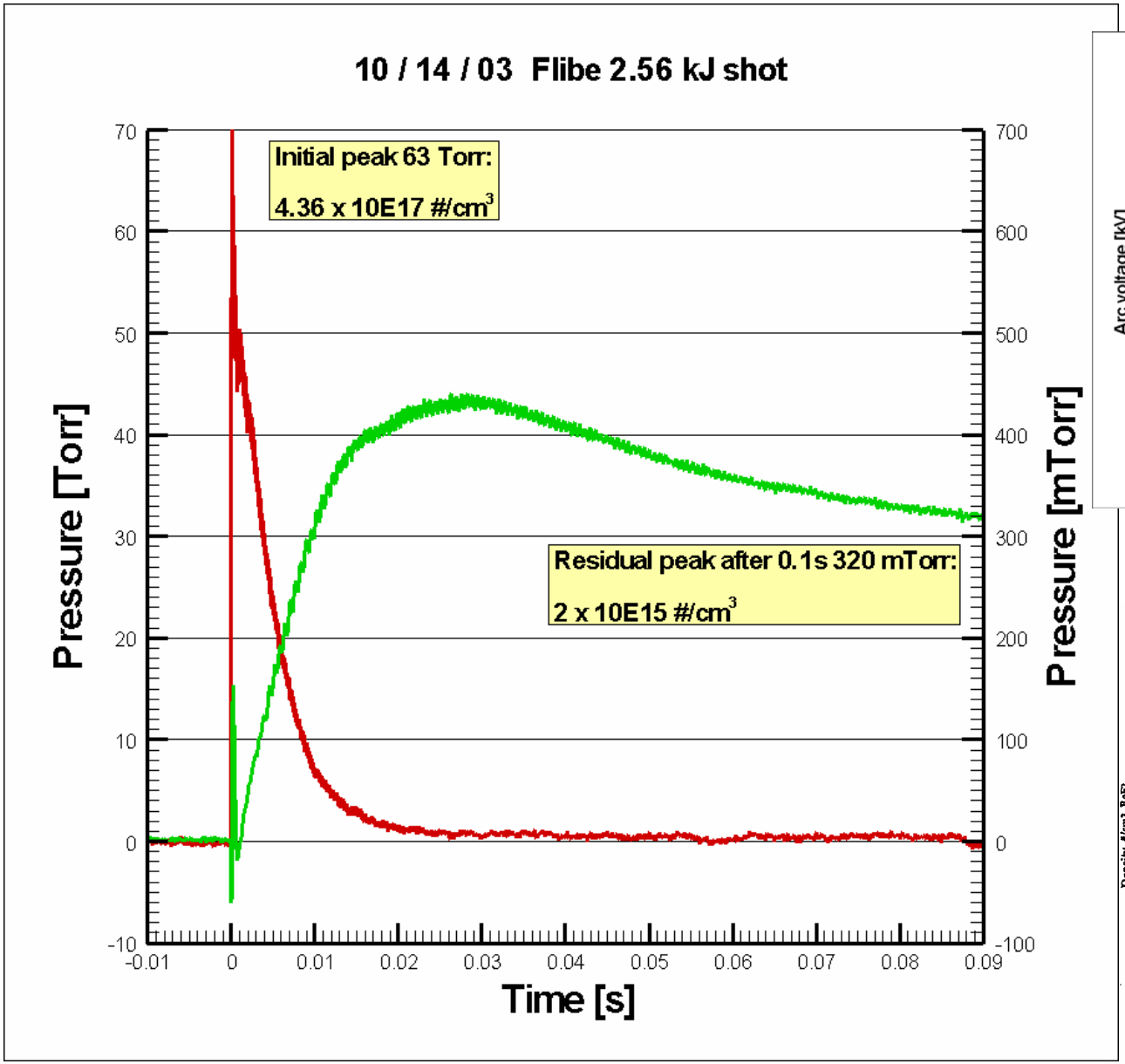
Cathode: 10'' W rod

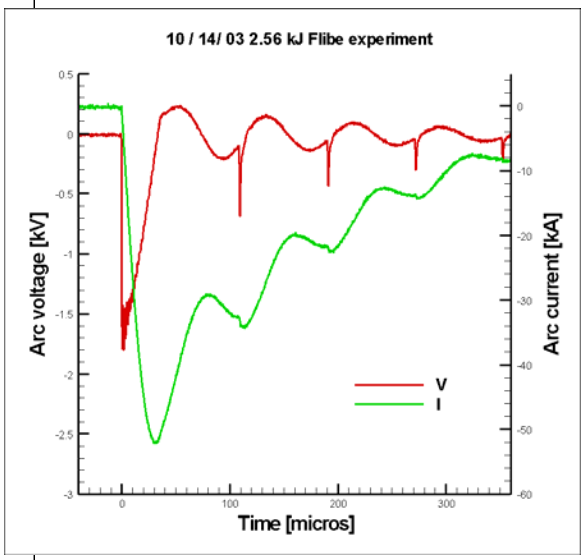
Flibe liquid pool

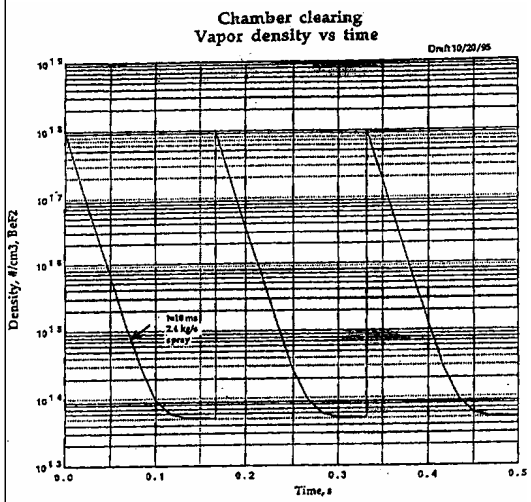
Anode: Nickel crucible with embedded high density cartridge heater, separately pumped to avoid oxidation of external surfaces

- High temperature compatible assembly - externally cooled pressure sensor sets upper limit (sensor < 260 C)
- E x B force traps plasma ions in the cavity above the liquid pool, increasing the confinement time and thus the amount of vaporized materials
- Flexible design for breakdown studies / liquid pocket implementation

Flibe experiments results







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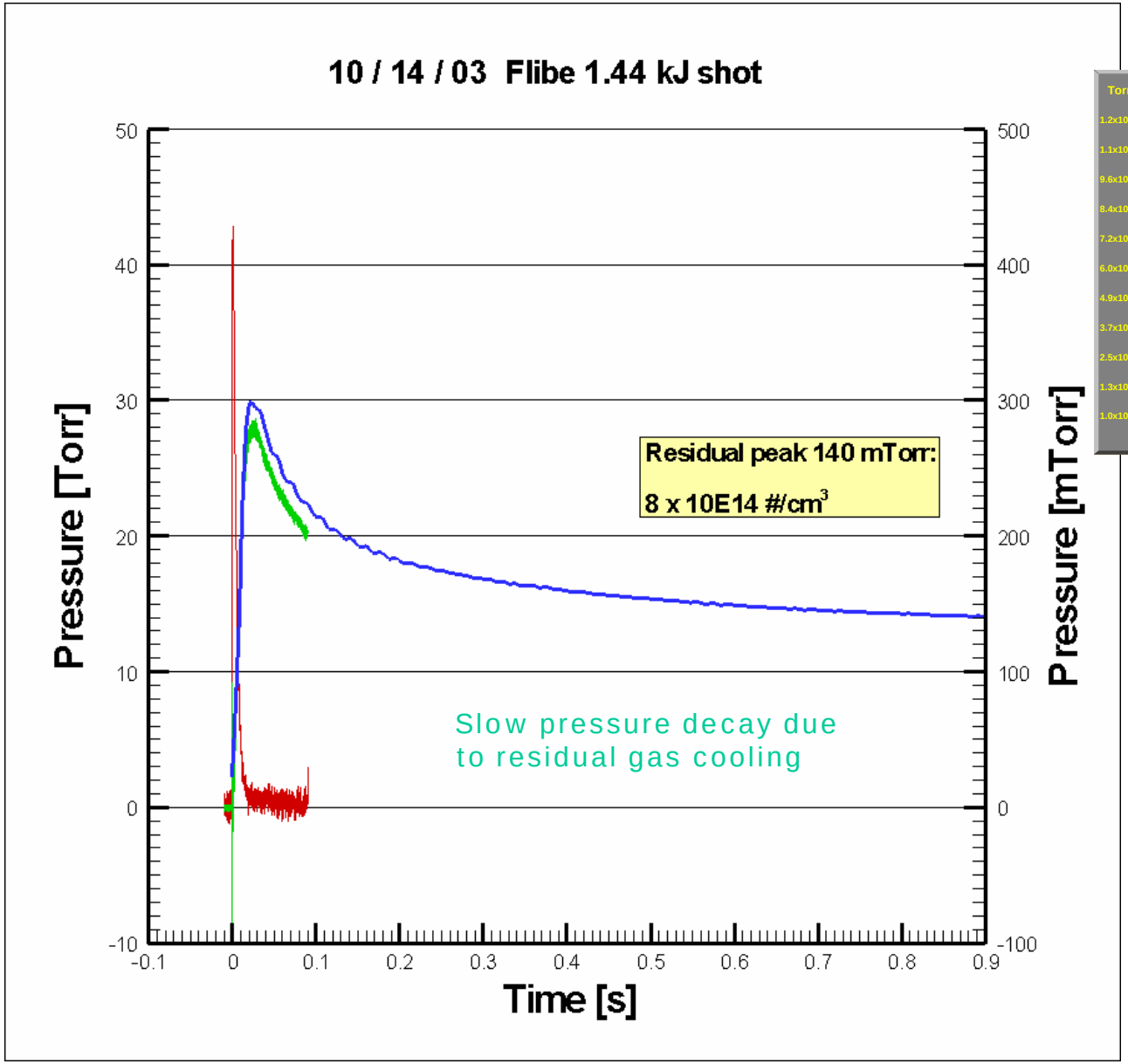
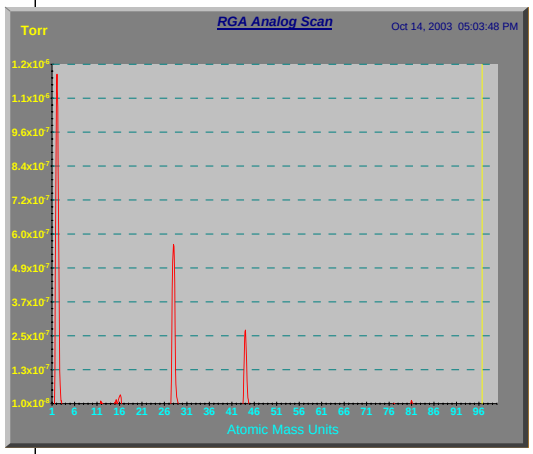
Flibe experiments results

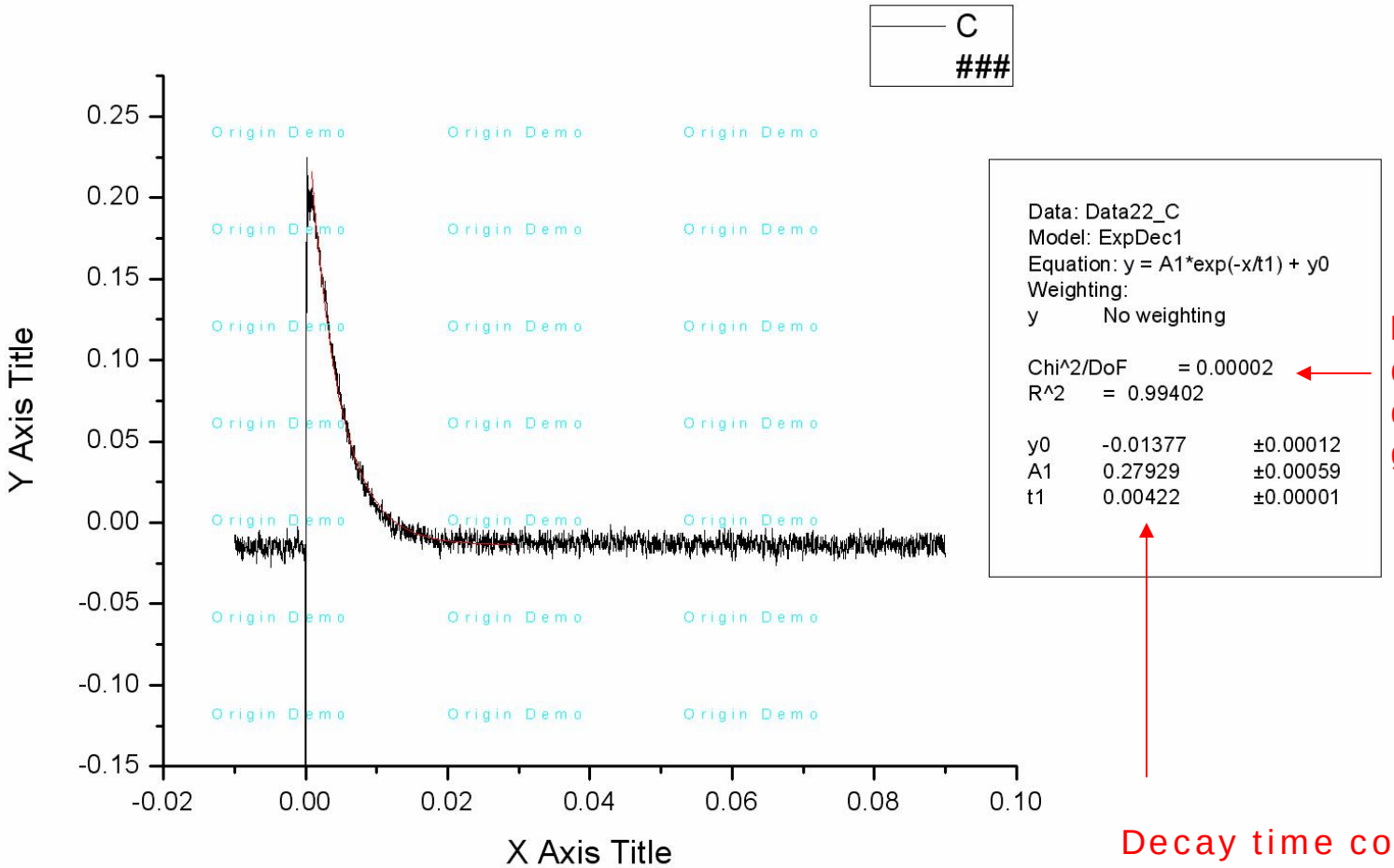




Residual gas from impurities contained in the flibe itself as received from INEEL, and due to handling and processing (irreversible):

H2
CO
CO2

Flibe experiments results

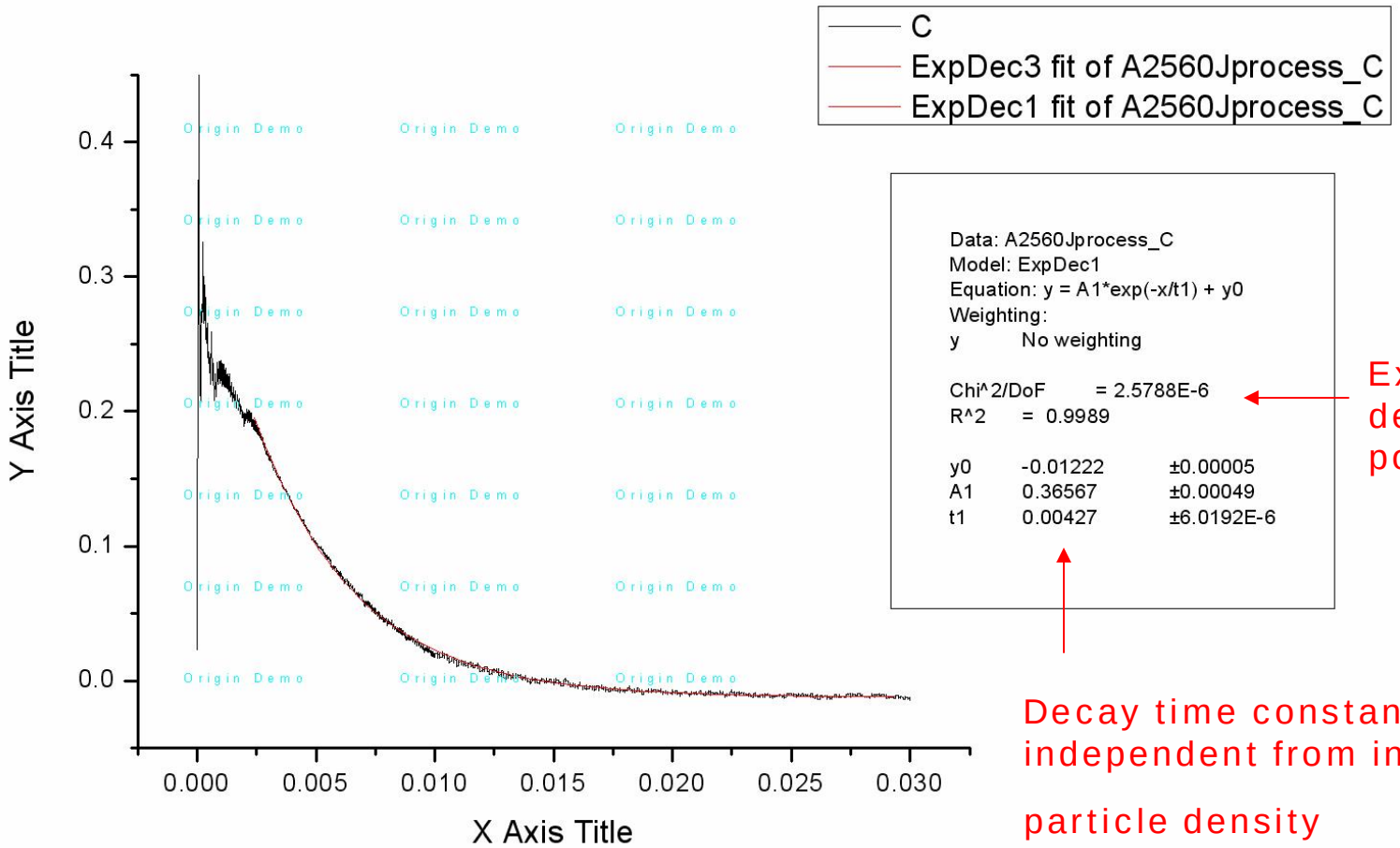


Exponential decay fits data points good

Decay time constant t_1 is a measure of the condensation rate

t_1 = 4.22 ms

Flibe experiments results



Exponential decay fits data points good

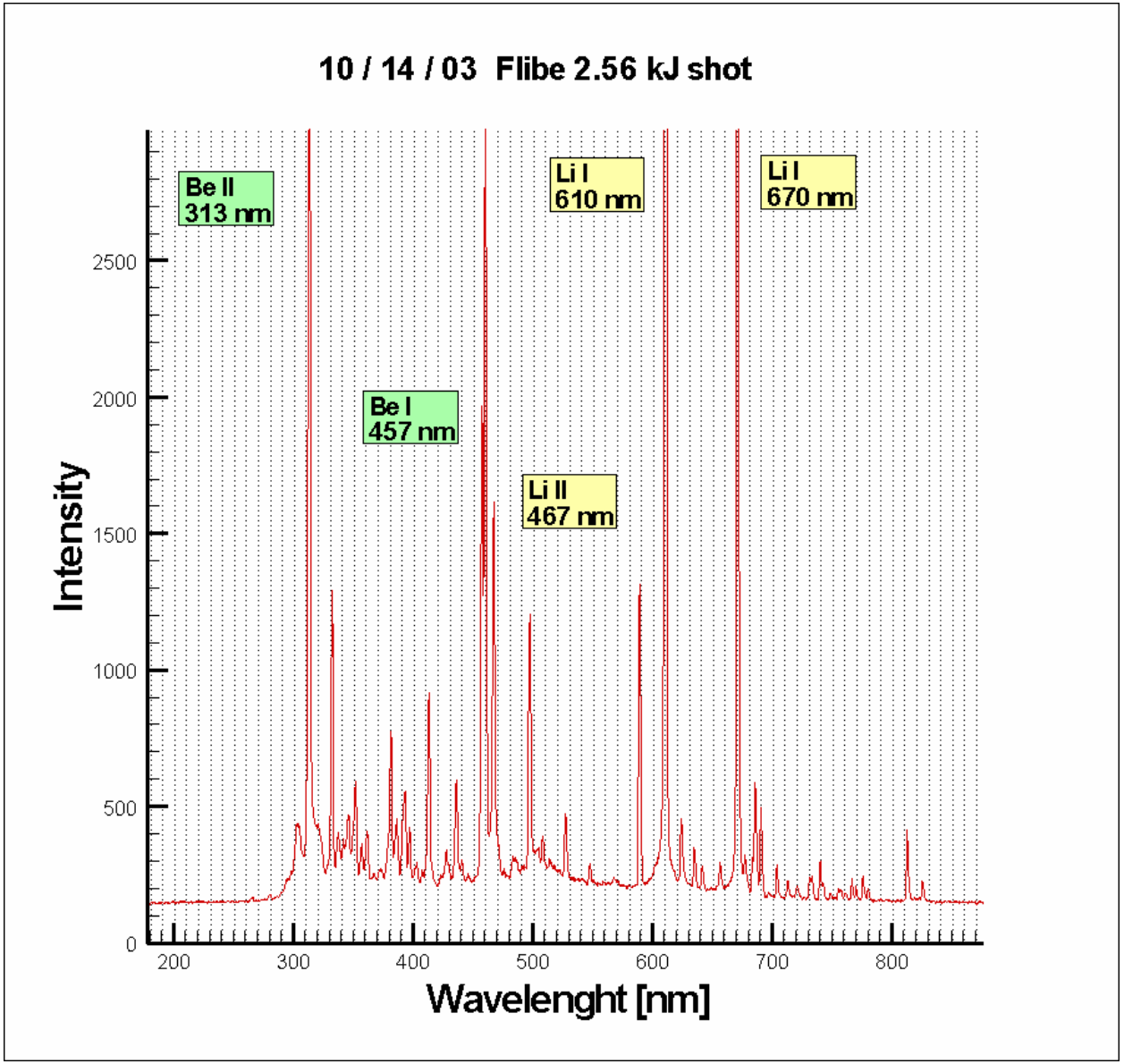
Decay time constant t_1 is independent from initial particle density

t_1 = 4.27 ms

Verify time constant for:

- higher initial particle density (discharge power)
- higher chamber wall temperature (partial results obtained)
- controlled background of non condensable impurities

Flibe experiments results



Emission spectroscopy:

strong lines from neutral and first ionization state of both lithium and beryllium atoms are present in the spectrum

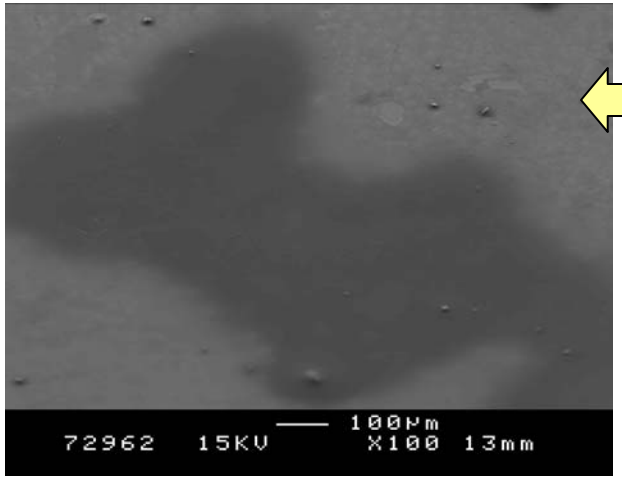
vapor characterization is possible after calibration

relative density of BeF_2 and LiF can be evaluated

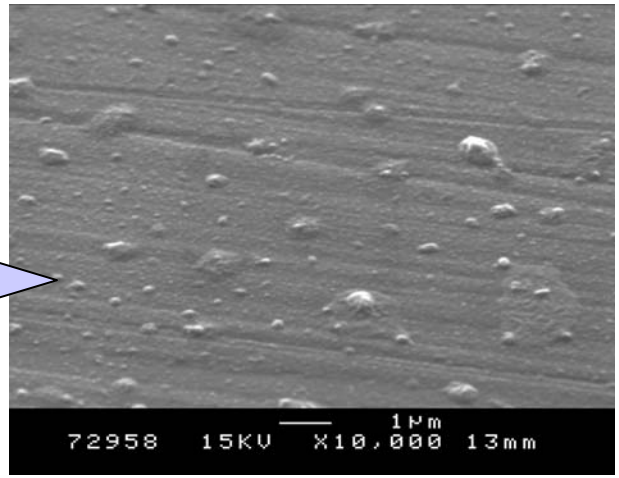
Flibe experiments results: SEM analysis

Vertical collecting plate (SS) perpendicular to the (radial) direction of vapor velocity - facing plate

Plates are coated with a gold film

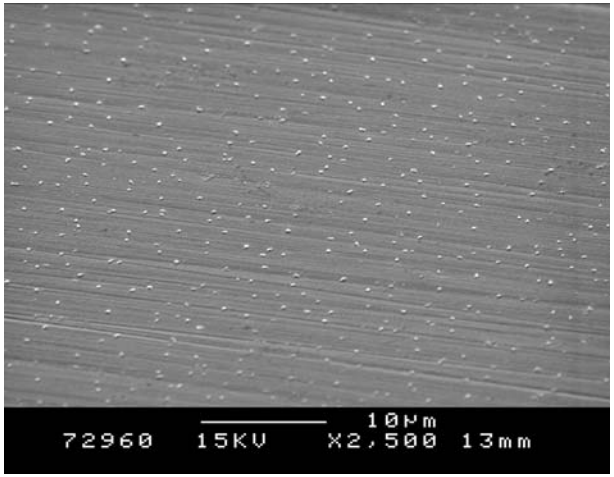


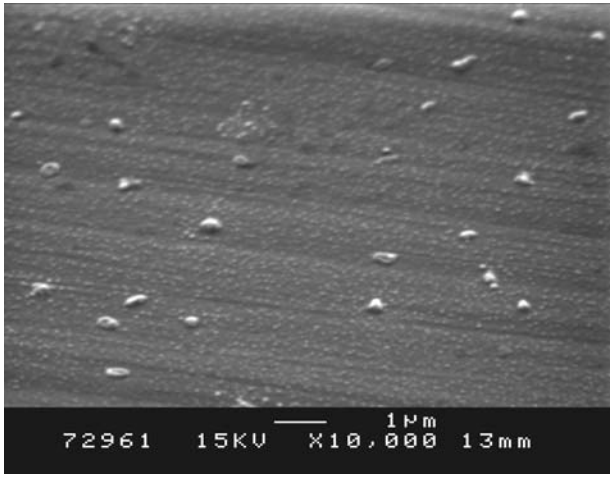
Large structures of surface vapor deposition - no evidence of droplet collection



Areas of non uniform, large droplets condensation or collection - 1 micron size

Areas of uniform, small droplets condensation or collection - 0.1 micron size





Flibe experiments results: SEM analysis

Vertical collecting plate (SS) parallel to the (radial) direction of vapor velocity - side plate

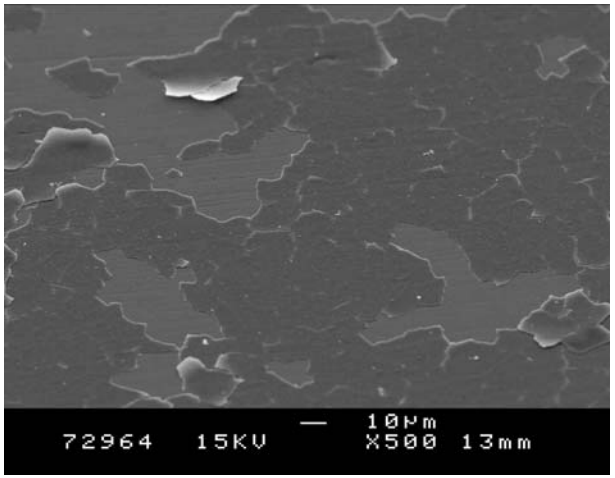
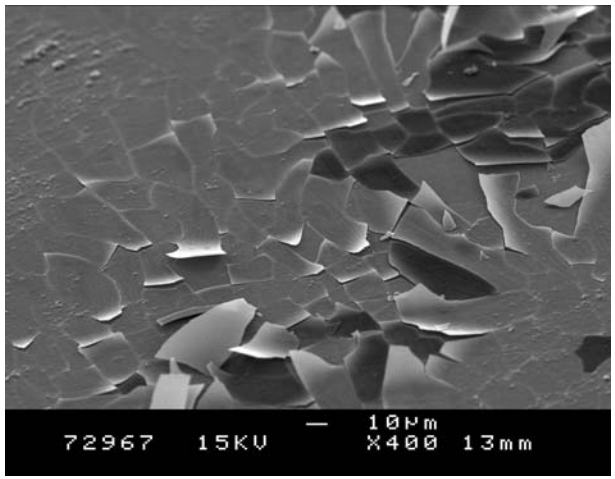
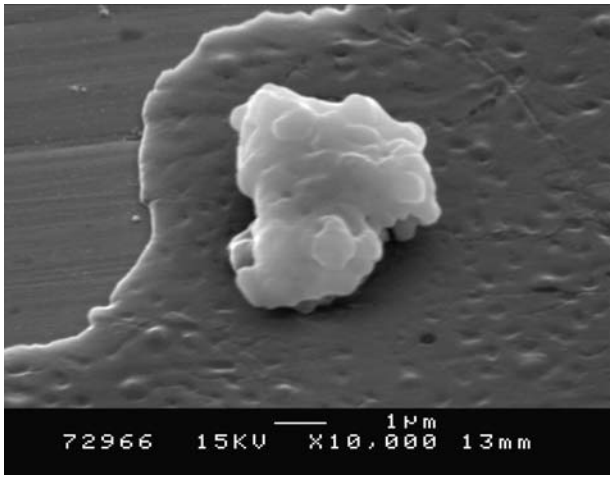
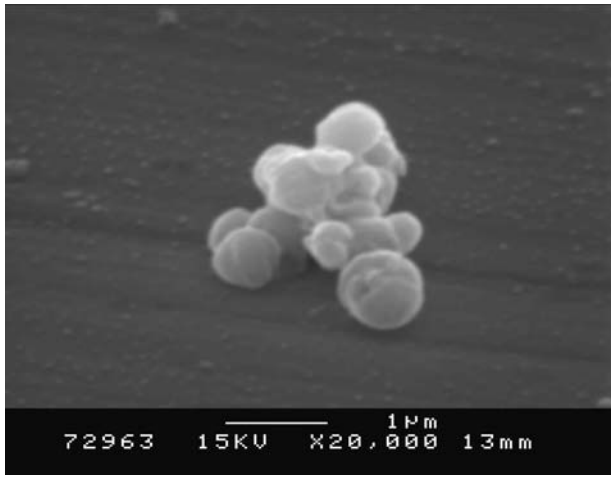


Plate is mostly covered by a condensate film that peels off the surface locally



On both plates there is unexpected evidence of particulate that is elctrostatically attached to the surface - health hazard





Modeling

Couple model for condensation at a liquid / vapor interface based on Schrage kinetic theory with UCB Tsunami calculations in 2-D asysimmetric volume

Apply enhanced Tsunami to simulate flibe vapor condensation experiments maintaining all assumptions implicit in Tsunami and condensation model

Compare with experiments
Evaluate effect of measured interface conditions:
traces of non condensable gases
vapor density dropping into transitions regime
accumulation of less volatile BeF2

Condensation model assumptions

2-D gas dynamics calculations assumptions:

- Two dimensional geometry
- Gas phase is continuum
- Gas state changes are isoentropic everywhere except at shock waves, treated as discontinuities
- Liquid structures are rigid - liquid inertia in the time scales of interest prevents the structures to move or deform - no work is transferred from gas to liquid
- Gas viscosity is negligible - viscous time scale L^2/ν >> dynamic time scale L/c
- In the volume gas is adiabatic - conduction time scale L^2/α >> dynamic time scale L/c - radiative losses assumed negligible
- Flibe is an ideal gas law with constant γ - fitted EOS corrected for dissociation and ionization not effective in the considered range

Condensation model assumptions

1-D liquid / vapor interface assumptions:

- Vapor condenses only on liquid surfaces present as initial condition - no droplets nucleation in the volume
- Liquid layers are semi-infinite slabs - thermal diffusion length $(\alpha\ \Delta t)^{1/2}$ << surface curvature - initial layer T is uniform - liquid T away from the interface remains constant - droplet spray cooling not considered
- Heat and mass transfer at the interface only in the normal direction - interface velocity due to mass addition is neglected because of mass continuity
- Heat transfer in the liquid layer by conduction in the normal direction - a convection term due to condensing flux introduced in the energy eq
- Liquid surface is always in thermodynamic equilibrium with the vapor - high mass transfer rates during initial transient neglected - continuum assumption
- Recombination and chemical diffusion effects for flibe is fast - vapor chemical composition is fixed by initial conditions
- Vapor composition is uniform in the volume and at the interface
- Interface kinetic theory accommodation coefficients (sticking and evaporation) are assumed to be = 1

Interface condensation: Schrage theory

The effect of condensation on the molecular motion is to impose a net flux in the direction normal to the interface:

$$\frac{dN_{u,v,w}}{N} = \left(\frac{m}{2\pi k_b T}\right)^{3/2} \exp\left[-\left(\frac{m}{2k_b T}\right)\left[(u-u_0)^2 + v^2 + w^2\right]\right] du\, dv\, dw$$

Integrating over v, w and positive (toward) u:

$$\phi_N^+ = n_0 \left(\frac{k_b T}{2\pi m}\right)^{1/2} \left\{ \exp\left[-u_0^2\left(\frac{m}{2k_b T}\right)\right] + u_0\sqrt{\pi}\left(\frac{m}{2k_b T}\right)^{1/2}\left[1 + erf\left(u_0\left(\frac{m}{2k_b T}\right)^{1/2}\right)\right]\right\}$$

Re-writing:

$$G^+ = p\left(\frac{\mu}{2\pi R_0 T}\right)^{1/2}\left\{e^{-s^2} + s\sqrt{\pi}\left[1 + erf(s)\right]\right\}$$

where

$$s = u_0\left(\frac{\mu}{2R_0 T}\right)^{1/2} = \frac{u_0}{(\gamma RT)^{1/2}}\left(\frac{\gamma}{2}\right)^{1/2} = M\left(\frac{\gamma}{2}\right)^{1/2}$$

$$p = \rho RT$$

Finally the net flux across the interface:

$$G_k = f_c\, p_{vs}\, \Gamma\left(\frac{1}{2\pi RT_{vs}}\right)^{1/2} - f_e\, p_{ls}\left(\frac{1}{2\pi RT_{ls}}\right)^{1/2}$$

Modeling the interface and liquid layer

Two equations to couple liquid and vapor properties at the interface:

mass balance $$G\big|_{x=0^-} = \rho_{vs}\, u_{vs} = \rho_{ls}\, u_{ls} = G\big|_{x=0^+} \qquad t \geq 0$$

energy balance $$G\, h_{vs}^0 = -k_l \left.\frac{\partial T(x,t)}{\partial x}\right|_{x=0} \qquad t \geq 0$$

conduction in the vapor neglected for short diffusion length - neglect radiation

Vapor stagnation enthalpy: $$h_{vs}^0 = c_{pg}\left[T_{vs} - T_{sat}(p_{vs})\right] + h_{fg}\left[T_{sat}(p_{vs})\right] + \frac{u_{vs}^2}{2}$$

Where for flibe

$$h_{fg}(T_{sat}(p_v)) = h_{fg}(T_{ref}) + (c_{pg} - c_{pl})\left[T_{sat}(p_v) - T_{ref}\right] \qquad p_{sat} = \frac{101300}{760} \times 10^{\left(9.407 - \frac{10054}{T}\right)}$$

In the liquid layer:

Energy equation $$\frac{\partial T(x,t)}{\partial t} + u_{ls}\frac{\partial T(x,t)}{\partial x} = \alpha \frac{\partial^2 T(x,t)}{\partial x^2} \qquad 0 \leq x \leq \infty$$

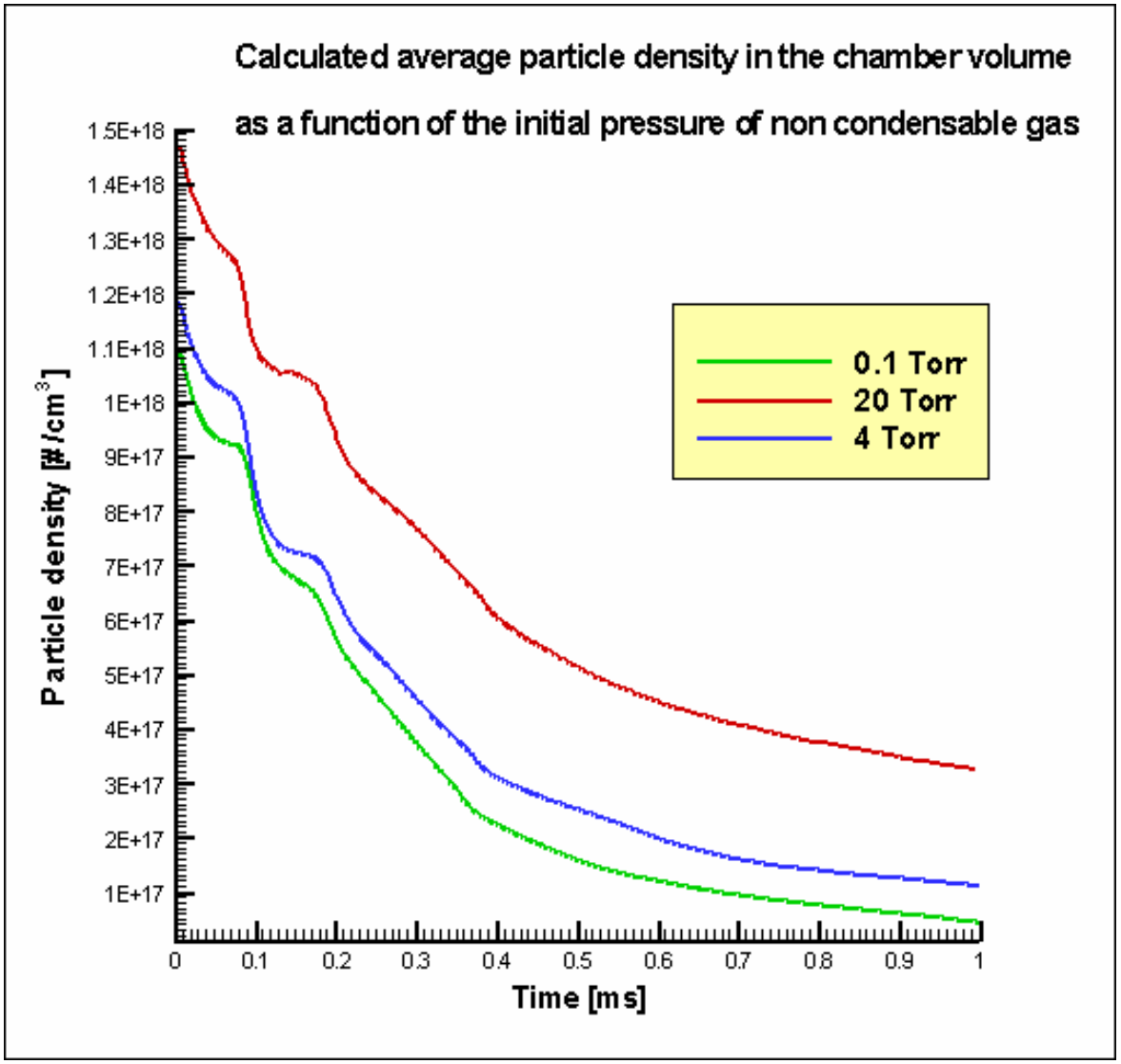
with bc

$$q''_{\text{int}} = -k_l \left.\frac{\partial T(x,t)}{\partial x}\right|_0 \qquad t \geq 0$$

$$T(\infty,t) = T_{l\infty} \qquad t \geq 0$$

Convection term added to account for condensing mass across the interface - u_{ls} evaluated from G

Additions to the condensation module: non-condensable component



Using Tsunami multi-component capability, the gas is simulated as a mixture of injected flibe vapors and background, non-condensable gas

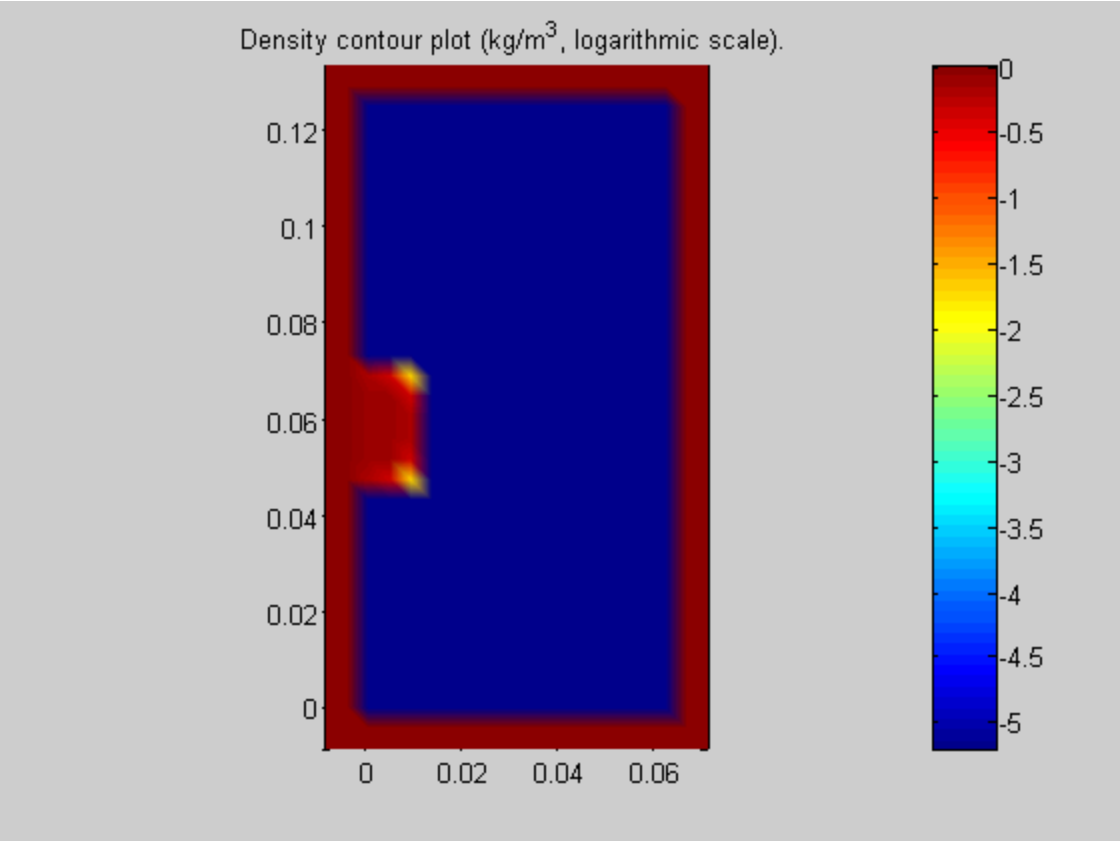
Injected gas is flibe vapor, all mass available in a 3 x 3 cell volume at the center of the numerical domain and at time 0. Properties are determined by its total initial energy and mass

Background gas properties are determined by initial pressure and temperature

Chemical diffusion effects and accumulation of the non-condensable component at the interface cannot be considered by Tsunami itself

A boundary layer effect for heat and mass transfer could be coupled with the code (?)

Additions to the condensation module: time dependent injection source

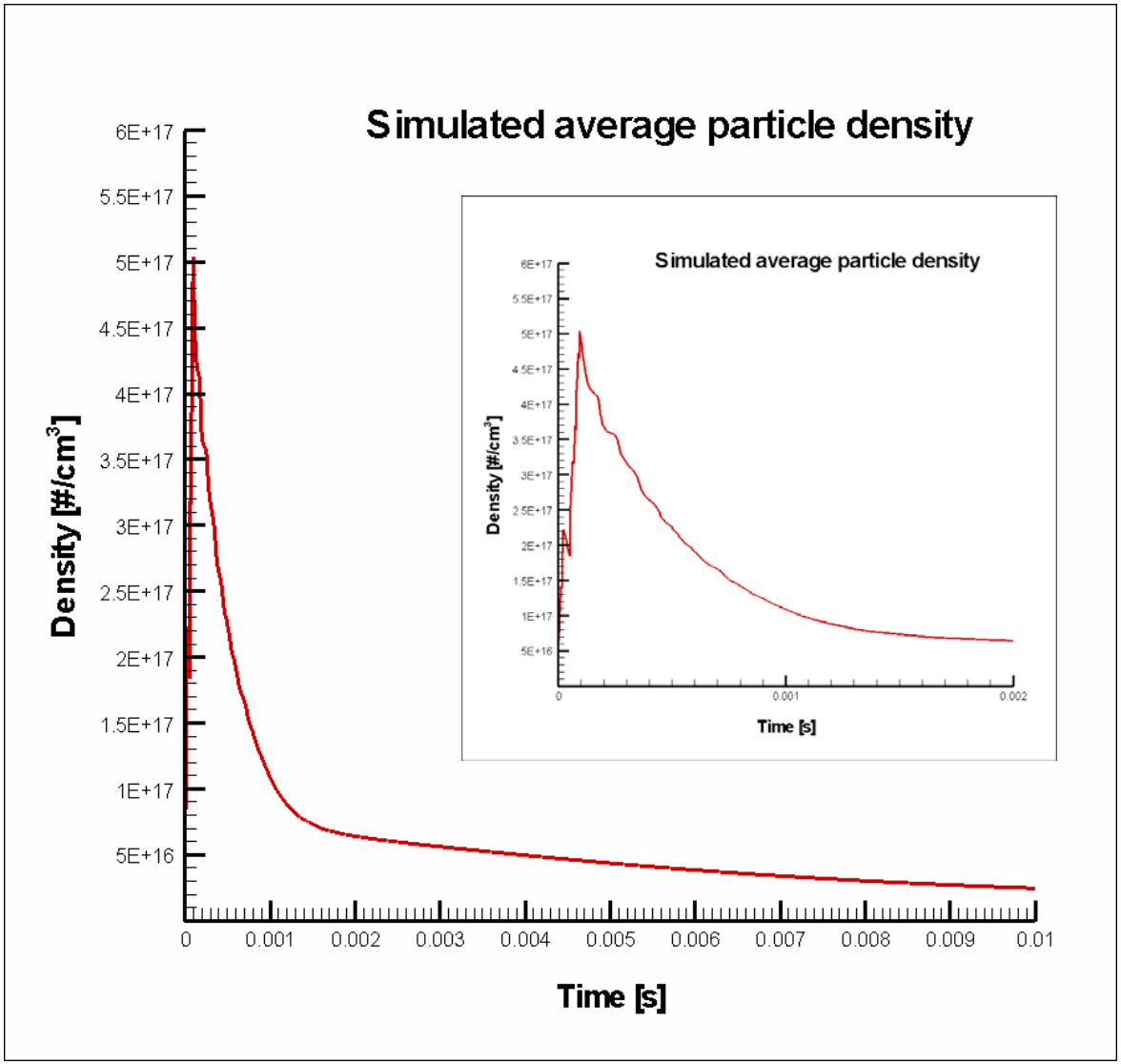


Tsunami calculations could not be extended beyond 1 ms because of problems in the convergence of the energy flux at the interface, where the boundary condition for mass, momentum and energy fluxes is determined by the condensation module

To avoid the high gradients due to the injection of all vapor mass at time 0, a time dependent constant source has been implemented in the code

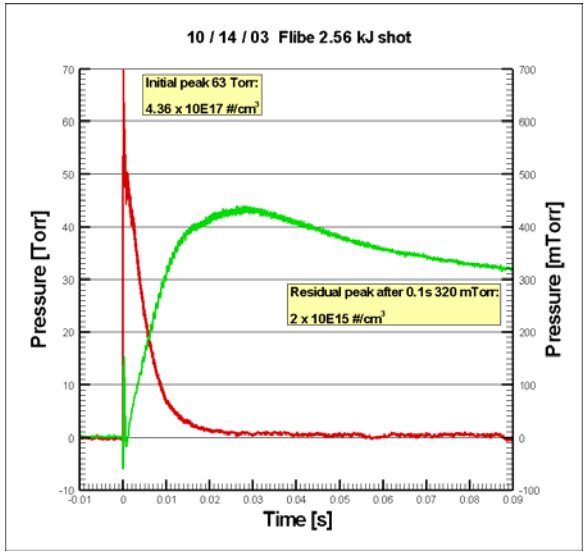
Movie shows density as a function of time as a sequence of contour plots.
Total duration of the sequence is 10 ms.

Preliminary comparison of numerical simulations with experimental data



Simulated time constant for condensation is one order of magnitude faster then experiments

The adiabatic assumption of Tsunami (no radiation, no heat transfer at the boundary) slows the condensation flux after the first ms



Summary

- Experimental characterization of the time constant for flibe vapor condensation will be extended to higher initial density and wall temperature, but first results show that the process is compatible with IFE power plant repetition rates

- However, first results also show that control of the impurities dissolved in the molten salt become a fundamental issue for design concepts (heavy ion beam drivers) that require recovery of vacuum conditions in the 10^{14} #/cm^3 range

- More detailed analysis of the condensation process in the low pressure regime can be performed with additional diagnostics that are currently under test and evaluation

- The Tsunami code with the condensation module overestimates the condensation rates by a factor of 10, and it's validity is limited to the initial few milliseconds of the condensation process. A boundary layer theory for heat and mass transfer needs to be included to extend its capability

Research coordination

Flibe condensation experiments can continue with focus on:

- impurities control in liquid flibe
- surface / volume condensation and particle size distribution characterization
- diagnostic development for vapor properties characterization

UCLA facility can be used for experimental investigation of other issues relevant to IFE HIB or laser chamber design

The End