Research and Development of a Solid-Stopper for the Facility for Rare Isotope Beams (SOL)

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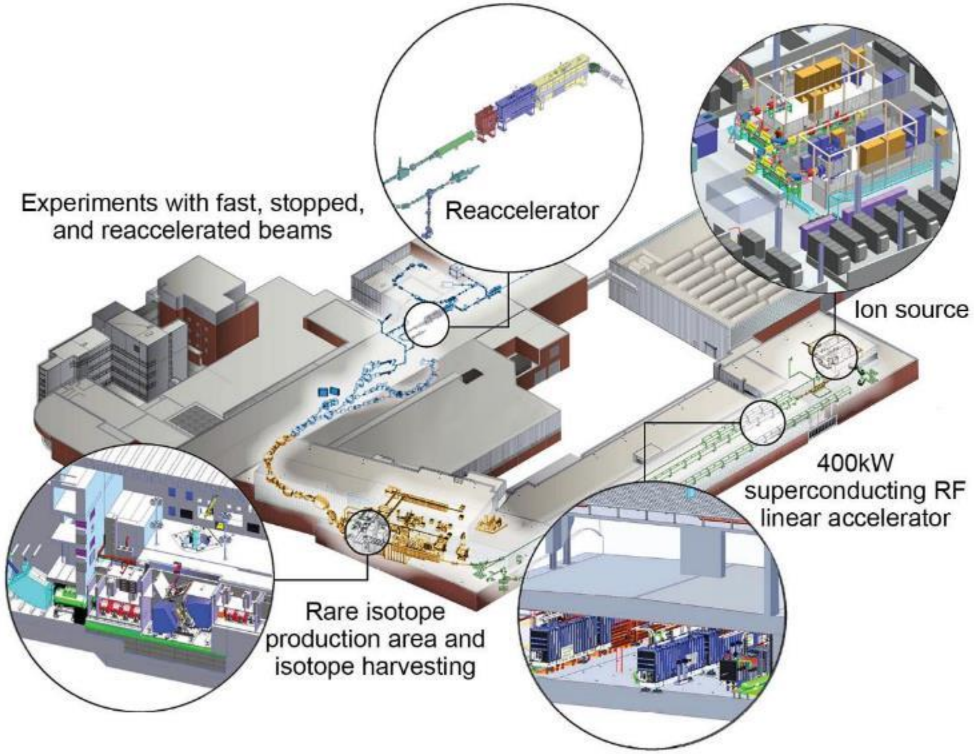
Outline

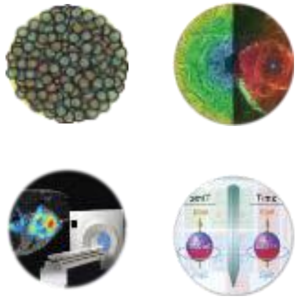
- FRIB provides fast, stopped and reaccelerated rare isotope beams for science
- Technique of stopped beams is well mastered at FRIB
- Gas stoppers are the first choice, however limitation on beam intensity and light ions need to be addressed
- Solid stoppers can complete the stopping scenario unlocking solutions for selected isotopes
  - FRIB opens opportunities to new material development
- SOL - Funding opportunity DE-FOA-0003261 status
  - Simulation of diffusion-effusion process with development with a code was performed
  - SOL design is well advanced
  - SOL temperature studies on-going
  - Schedule is defined

FRIB has Wide Range of Beams and Energies for Science

- Rare isotope production via in-flight technique with primary beams up to 400 kW, 200 MeV/u uranium
  - Presently 20 kW – on the road to achieve full power
- Fast, stopped, and re-accelerated beam capabilities
- Experimental areas and equipment for science for fast, stopped, and reaccelerated rare isotope beams

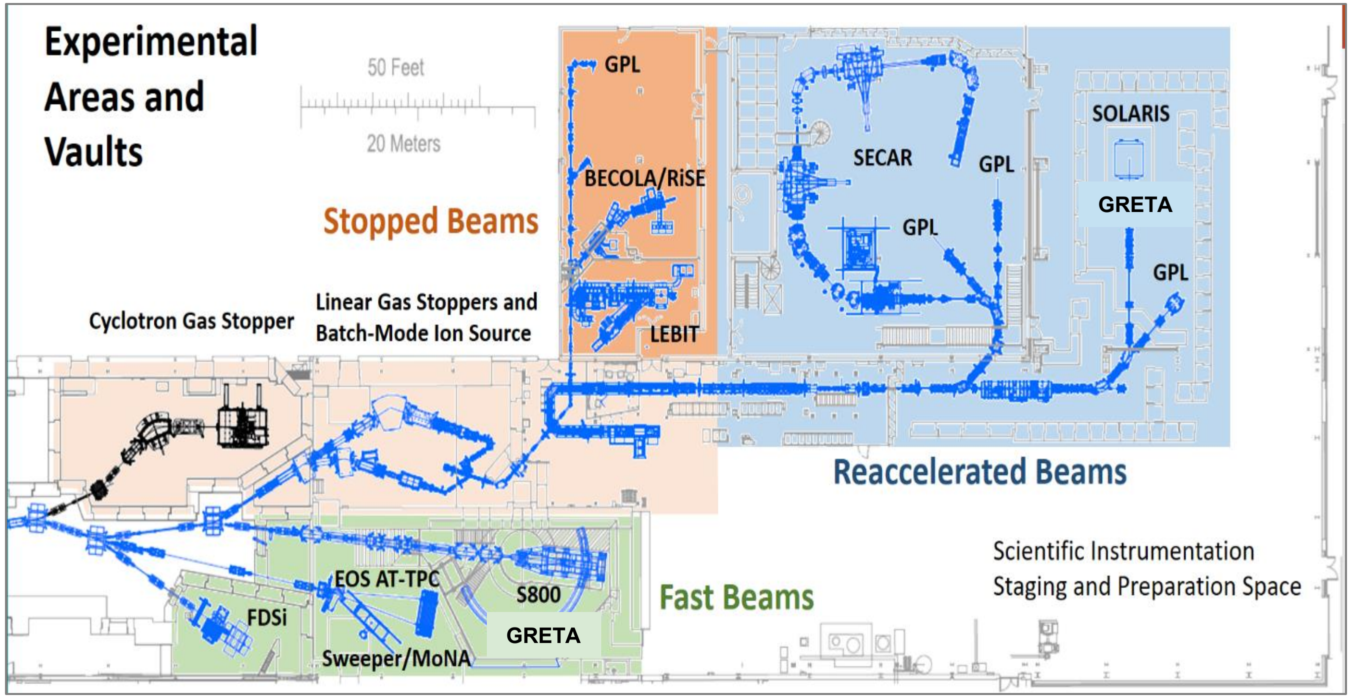


- **FRIB Science**
  - Properties of atomic nuclei
  - Nuclear astrophysics: What happens inside stars?
  - Tests of laws of nature
  - Societal applications and benefits



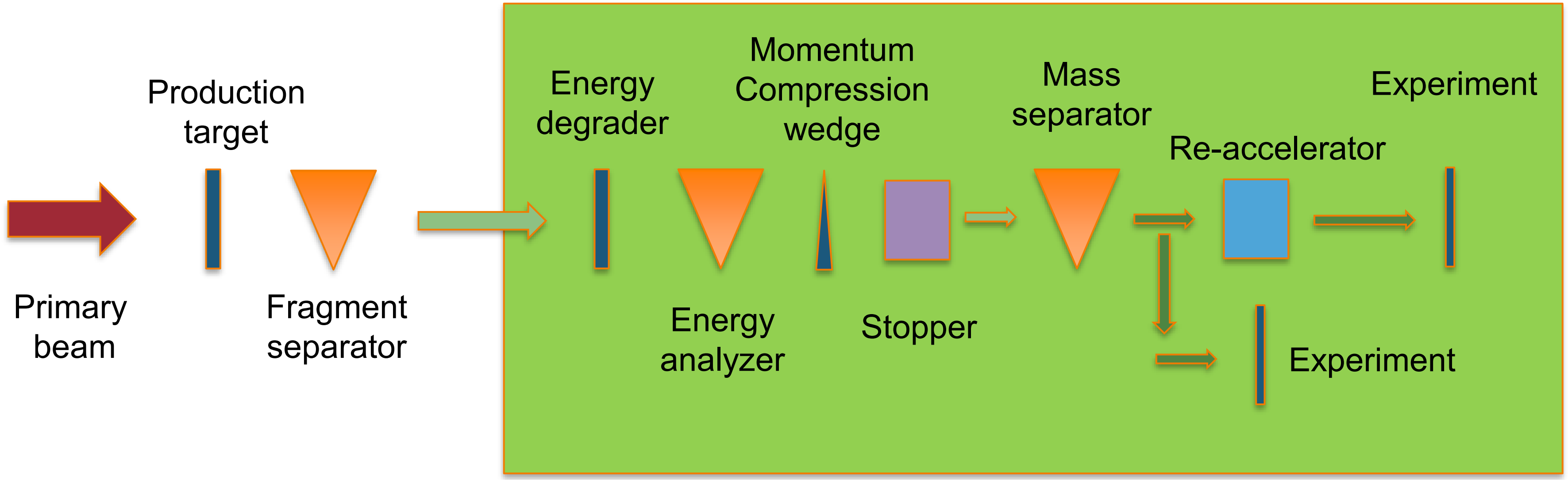
FRIB Provides Stopped and Reaccelerated Beams for Science, Making Use of a Variety of Instruments

- Stopped beams areas Instruments
  - LEBIT: mass measurements
  - BECOLA/RISE: charge radius, electromagnetic moments and nuclear spins
  - GPL: Two General purpose beamlines (for user's set-ups)
- Reaccelerated beams areas Instruments
  - SECAR: Separator for capture reactions
  - SOLARIS: Solenoidal spectrometer with
    - » AT-TPC
    - » Si-array
  - GRETA: Gamma-ray Energy Tracking Array
  - GPL (for user's set-ups)
    - » Total of 3 general purpose beamlines
- All those instruments take advantage of the gas stoppers in N4 vault or the Batch Mode Ion Source (BMIS)



Stopping Technique With or Without Reacceleration is Well Mastered in FRIB

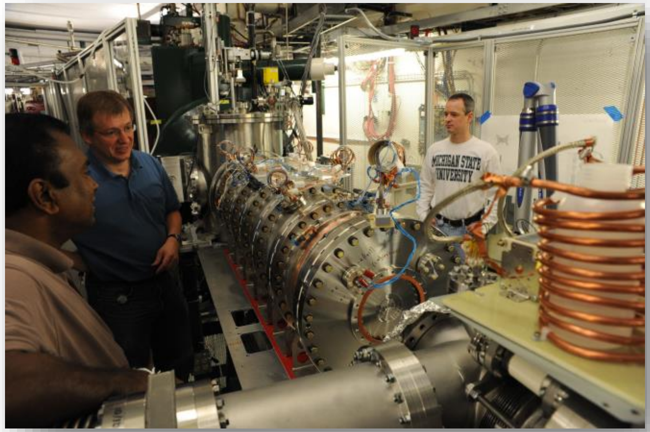
- The stopping technique is shown below
  - Note that FRIB is unique in stopping rare isotope fast beams and reaccelerating



- Stoppers can be gaseous or solid
  - For gas, helium is of choice, due to its high first ionization energy
  - For solid, refractory material is of choice, as high temperature must be used to extract implanted rare isotopes from the material matrix

FRIB Has Developed Successful Gas-stoppers
Two Presently in Operation and More to Come

- Room Temperature Gas Cell (RTGC) – N4 vault
  - Virtually can be used for all elements with exceptions of H, He
  - Optimal efficiency for half-lives down to about 200 ms
  - Provides overall efficiency (although fractionated due to molecular formation) between 5% - 30% for beam incident < 1E+7 pps
    - » Lower efficiencies for light ions Z<8



- Advanced Cryogenic Gas Stopper (ACGS) – N4 vault
  - Same as RTGC, with enhanced efficiency for a single mass, due to less fractionation (due to working at cryogenic temperatures, providing less contamination in the cell)
  - Optimal efficiency for half-lives down to about 100 ms (faster surfing technique)
  - For incident beam < 1E+8 pps provides overall efficiency between 5% - 30% (due to fast charge collection geometry)
    - » Lower efficiencies for light ions Z<8

Solid-Stopper Development Aims Mainly High-Intensity and Light Mass Stopped Beams

- Why a Solid-Stopper is needed
  - Gas-stoppers, in some cases, are arriving to their limits
    - » FRIB fast beam intensities **for isotopes close to the stability** are close to exceed present gas stopper capabilities, as beam yields in the gas cells can be > 1E+8 pps
      - This situation will worsen when FRIB would reach 400kW
    - » Cocktail beams provided by ARIS (mostly isotones) adds to the total charge implanted in the gas stoppers
  - Very light ions have poor efficiency in gas-stoppers and, in particular, noble gases

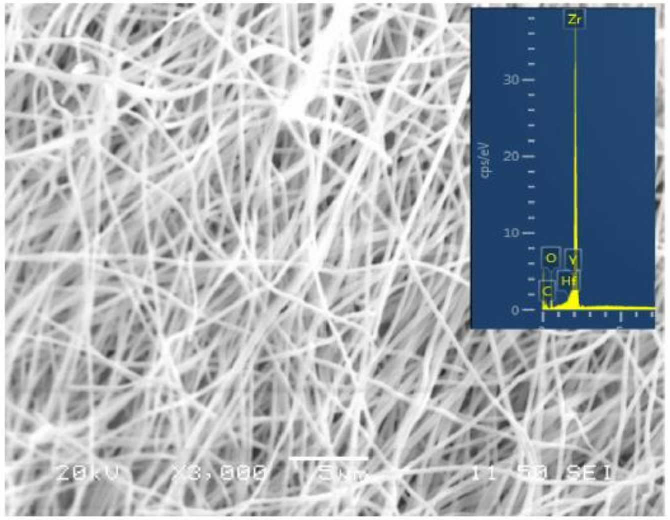
- Solid-Stopper advantages and disadvantages
  - Advantages:
    - » Virtually no fast beam intensity limitation (including cocktail beams)
    - » No light ion or noble gas limitation (solid stopper is even better for noble gases)
  - Disadvantages:
    - » Chemistry limitation due to diffusion and effusion to extract from the stopper and transport to the ionizer
    - » Need to re-ionize the isotope

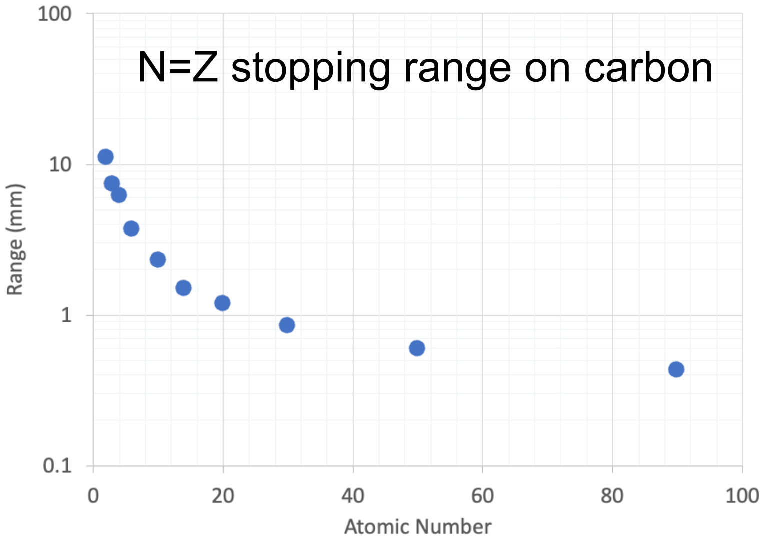
Solid-Stopper Technology Based on Available ISOL Development

- Solid-stopper technology is based on decades of Isotope Separation On-Line (ISOL) development; example of facilities:
  - CERN-ISOLDE
  - ISAC-TRIUMF
  - SPIRAL-GANIL
- Release, transport and ionization of rare isotopes from solid materials are well studied in ISOL facilities
- The difference from ISOL is that rare-isotope ions are not produced in situ; they are produced by in-flight fragmentation or fission and after, stopped and released from the stopper
  - The separation between production and release provides more flexibility, as the most resilient stopper can be used in all cases, independently from the production mechanism
- Techniques developed in ISOL as introduction of a reactive gas to form a molecule that releases easily, e.g., fluorination or oxidation, can be used
- Ion sources developed for ISOL e.g., Surface ionization (SI), Resonance Ionization Laser Ion Source (RILIS), Forced Electron Beam Induced Arc Discharge (FEBIAD) source or Electron Cyclotron Resonance (ECR) ion source can be used

FRIB Opens Opportunities to New Material Development for Solid-stoppers

- The stopping range of FRIB's fast beams in solid material is short (some millimeters). This allows thin stoppers to be used with the prospect of short release times.
  - The range of Z = N ions at 50 MeV/u after passing a degrader is shown, as a function of atomic number in a carbon stopper.
- As the range is short, new materials with low density could be used
  - Nanomaterials available in industry
    - » Carbon nanomaterial
      - Nanotube, quantum dots, nanofibers
    - » Oxides
      - Graphene oxide
      - Al_2O_3, MgO, etc. nanoparticles
      - Nanofibers





SEM image of zirconia nanofiber (FNAL courtesy)

FRIB
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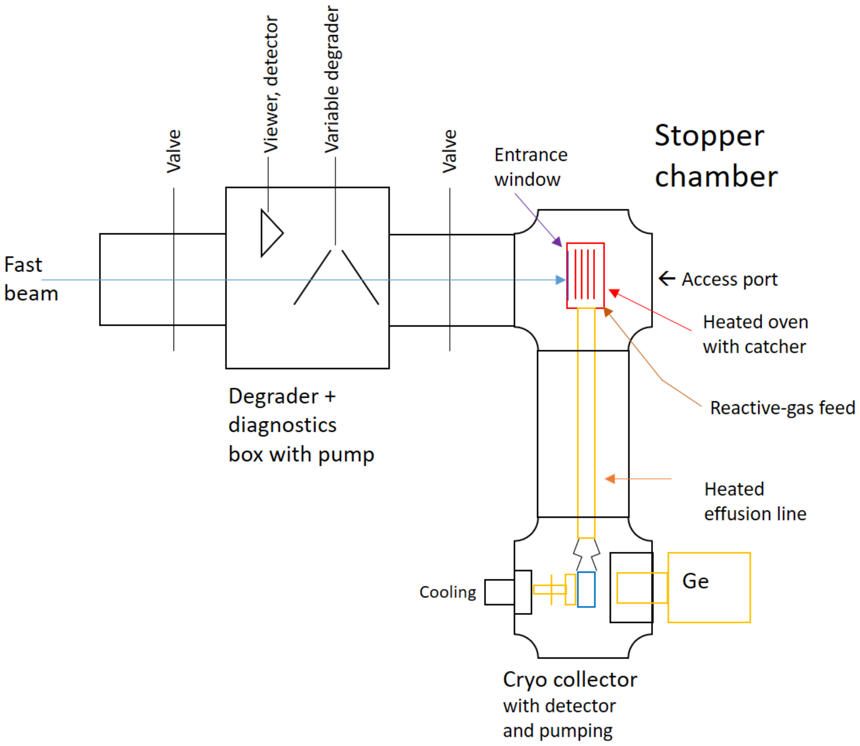
SOL Goals

- SOL: Research and Development of a Solid-Stopper for the Facility for Rare Isotope Beams
  - SOL focuses on studying the release efficiency of rare isotopes stopped in solid stoppers.
  - When combined with an ion source in the future, this concept will enable beam rates of rare isotopes for certain elements to surpass those achievable with gas stoppers by orders of magnitude.
  - The combination of fast-beam rare-isotope production, as conducted at FRIB, with a solid-stopper ion-source technique has never been applied anywhere worldwide.
  - Goals:
    - » Design and build a prototype system, which can accommodate solid stoppers of various kinds, including microstructure and nanostructured materials, at temperatures up to 2,300 K for efficient release by diffusion.
    - » Design and build a cryo-catcher connected to the solid-stopper for collecting rare isotopes released from the stopper. A HPGe detector will detect the characteristic gamma-ray of the various isotope decays.
    - » Perform irradiation of solid stoppers made of different materials and geometries with different rare-isotope beams and measure the release properties.

Concept of SOL Allows Testing Stoppers With Various Isotopes

- Fast beams from ARIS impinge the stopper in a chamber at 2,300 K
  - Fast beam intensity is measured by a plastic scintillator or directly by gamma rays from decay of isotopes using a HPGe detector, with the stopper cold (Ni)
- Isotopes are released and transferred to a cryo-collector via a heated effusion line
- Gamma rays from the decay of isotopes are detected by HPGe calibrated detector (Nf)
- The ratio Nf/Ni provides the efficiency of diffusion and effusion of the stopped isotope at a certain temperature



Processes During Release: Diffusion – Release Efficiency out of the Production Target

- We consider diffusion independent of other processes
  - Diffusion is the process of isotope releasing from the target matrix
- Diffusion is governed by Fick's law
  - Supposing a homogeneous distribution of the isotopes in the target matrix with thickness d, the delay function is:

$$p_\mu(t) = \frac{8\mu_0}{\pi^2}\sum_{n=0}^{\infty} e^{-\mu_n t}$$

  - Where: $\mu_n = \mu_0(2n+1)^2$ $\quad \mu_0 = \pi^2 D/d^2$ $\quad D = D_0 e^{(-E_A/kT)}$

μ_0 is the delay parameter and E_A and D_0 the Arrhenius coefficients and $\lambda = ln(2)/T_{1/2}$

  - Results:

$$\xi_d = \frac{tanh\sqrt{\lambda\pi^2/4\mu_0}}{\sqrt{\lambda\pi^2/4\mu_0}}$$

foil

$$\xi_d = \frac{3(\sqrt{\pi^2\lambda/\mu_0}\coth\sqrt{\pi^2\lambda/\mu_0} - 1)}{\pi^2(\lambda/\mu_0)}$$

sphere

R. Kirchner, Nucl. Instr. & Meth. in Phys. Res. B70 (1992) 186
M. Fujioka and Y. Arai, Nucl. Instr. & Meth. in Phys. Res. 186 (1981) 409

Processes During Release: Effusion – Transport Between the Target and the Ion Source

- The delay by effusion is in analogy of the evacuation time through an orifice

$$p_\nu(t) = \nu e^{-\nu t}$$

Where ν is the characteristic time of effusion

- The mean delay time can be written as

$$\tau_\nu^{del} = 1/\nu = \chi(\tau_a + \tau_f)$$

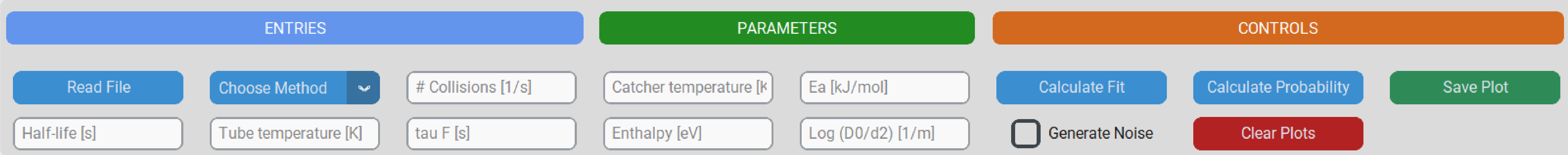
- Where χ is the average number of collisions on the walls of the transfer tube before reaching the ion source
- $\tau_{a,f}$ are the mean sticking time and the mean flight time between collisions
  - » Note that atoms, which hit the transfer tube will be absorbed and be re-emitted by the surface

- Result:

$$\xi_\nu = \nu/(\nu + \lambda) \quad ; \quad \tau_a = 2.4\ 10^{-15} e^{11650 \Delta H_a / T}$$

Diffusion-Effusion App Developed

- We have developed a Python script that allows for fitting the Arrhenius coefficients and enthalpy of adsorption.



- The code imports a CSV file with the measurements of temperature and yield.
- Data is fitted to the total yield (effusion times diffusion). $Y = y(\alpha) \cdot Y_\nu(\tau_{1/2})$
- Fit happens in two steps: global (Python Scipy Optimize differential_evolution) and local (Python Scipy Optimize curve fit).

$$Y_\nu(\tau_{1/2}) = \frac{\nu}{\nu + \lambda}$$

in which ν is the inverse of the mean delay:

$$\tau_\nu^{\mathrm{del}} = \frac{1}{\nu} = \chi(\tau_a + \tau_f)$$

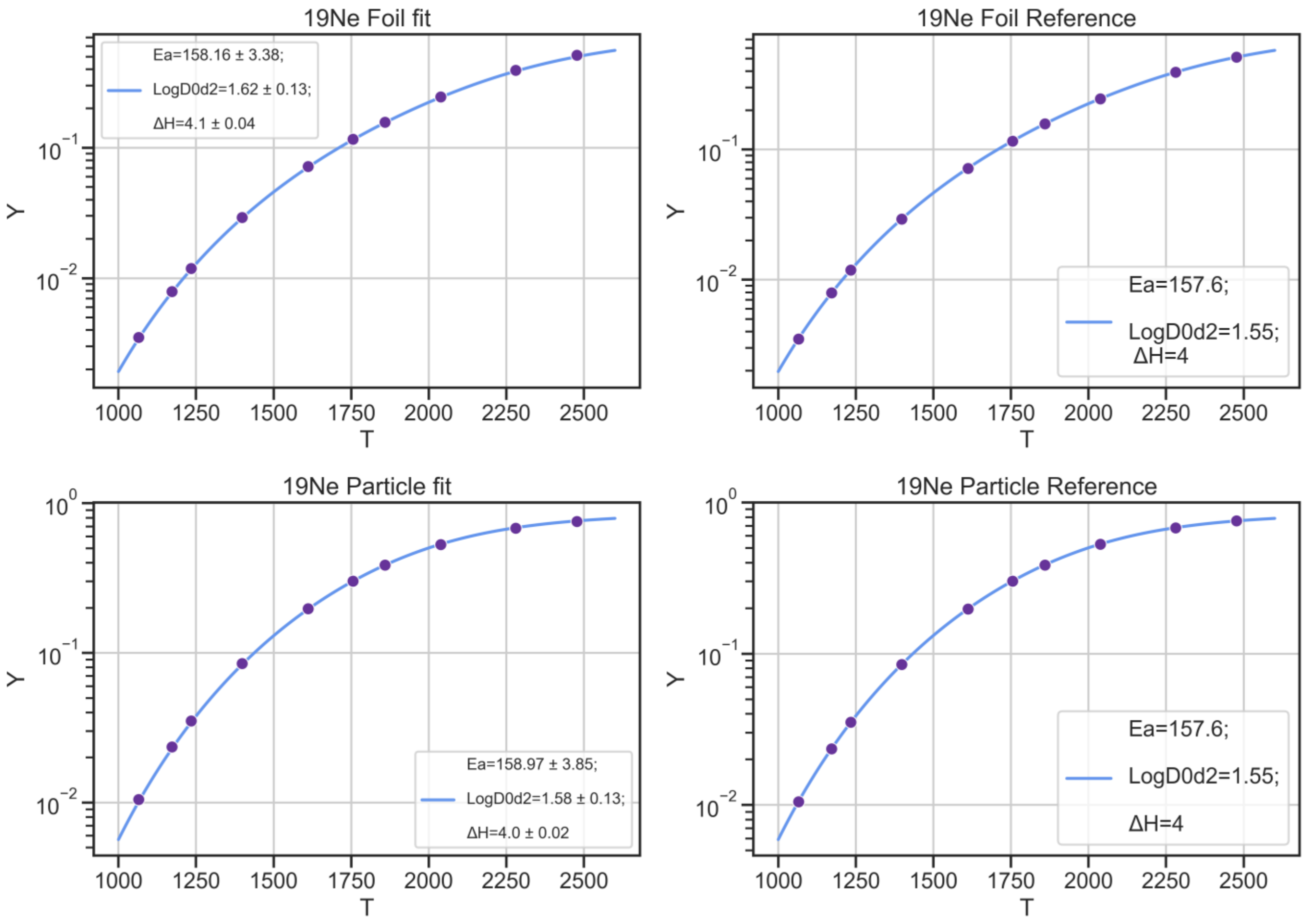
$$\alpha = \tau_N \cdot \frac{D_0}{a^2} \cdot \exp\left\{\left(\frac{-E_a}{R \cdot T}\right)\right\}.$$

$$y(\alpha) = \begin{cases} n = 1 : \alpha^{1/2} \cdot \tanh\left(\alpha^{-1/2}\right) & \text{Foil} \\ n = 2 : 2\alpha^{1/2} \cdot I_1\left(\alpha^{-1/2}\right) / I_0\left(\alpha^{-1/2}\right) & \text{Fiber} \\ n = 3 : 3\alpha^{1/2}\left[\coth\left(\alpha^{-1/2}\right) - \alpha^{1/2}\right] & \text{Particle} \end{cases}$$

Diffusion-Effusion App Results Validate the Method

- Example with simulated data and measured values.
  - Data is generated using equations sh

Parameters from N. Lecesne et al, Nucl. Intr. Meth in Phys. Res. B126 (1997) 141

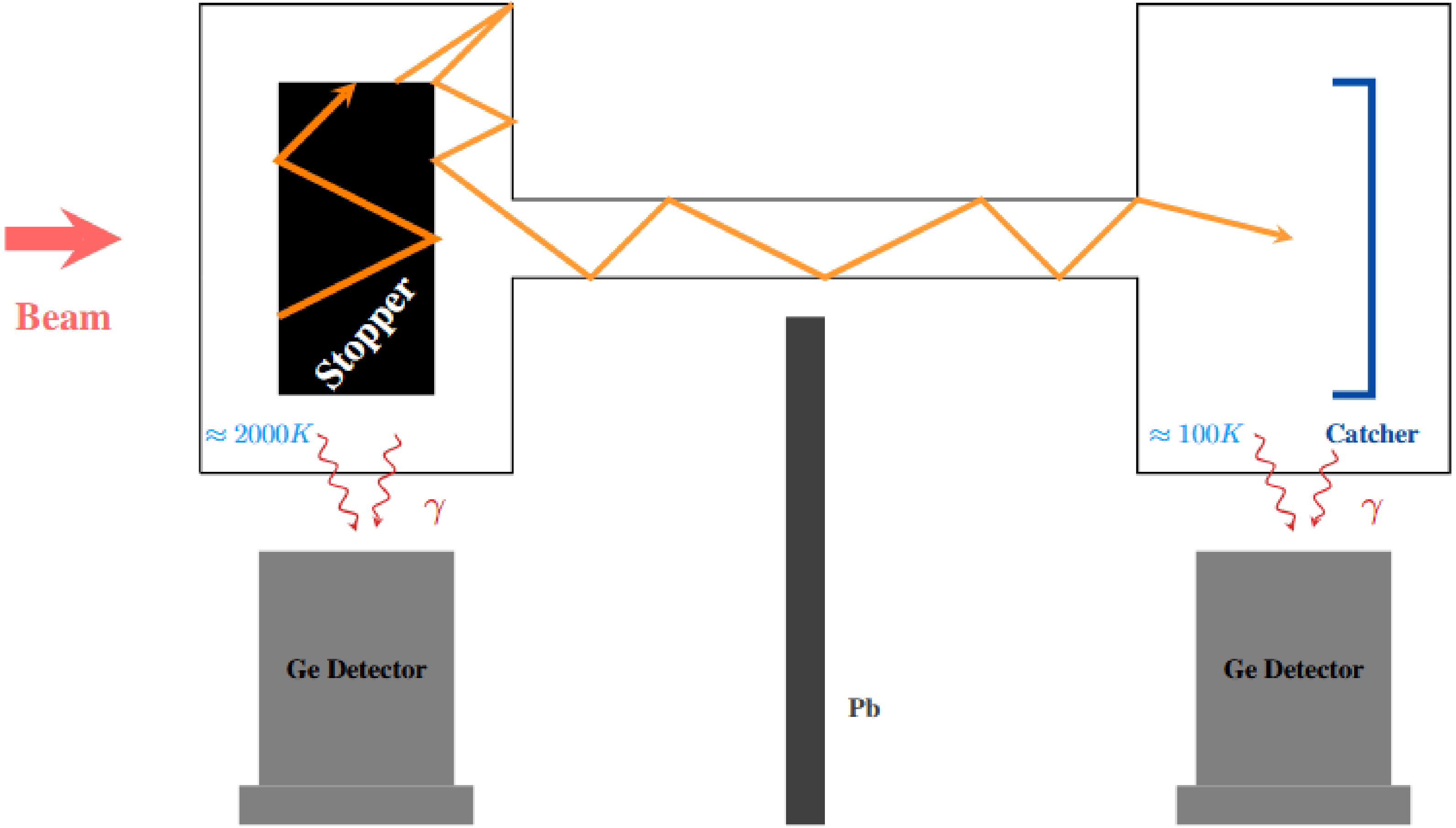


- Known values
  - Ea = 157.6 kJ/mol
  - Log(D0/d^2) = 1.55 1/m
  - Enthalpy = 4 eV

- Foil Fit values for 19Ne
  - Ea = 158.16 ± 3.38 kJ/mol
  - Log(D0/d^2) = 1.62 ± 0.13 1/m
  - Enthalpy = 4.1 ± 0.04 eV

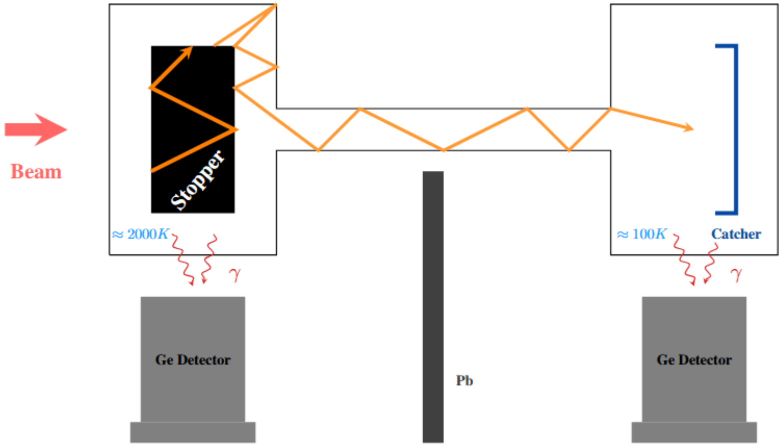
Experiment Proposal: Simulations for SOL

- Example of production and calculations.
  - 35Ar
- Simulation results.



Total Count Rate on Implantation Realistic

- Total efficiency for gamma rays (1.2 MeV)
  - = 1.35% x 5% x 0.1% x 87%
  - => 5.9E-7 (absolute)
- Total count rate on implantation: = 1E+9 x 5.9E-7 = 600 cps
  - For 10,000 counts (1% statistics) = > need 15 s
    - » This is in the implantation point
- Supposing Diff-Eff efficiency of 0.5% (lower temperature case)
  - For 1,100 counts (3% statistics) => need 6 min in the catcher



List of Isotopes To be Used With SOL

- 36Ar can be used to produce 35Ar, 34Ar, 32Cl, 31S, 16N, 15O, 14O, 13N, 8He.
- LISE++ initial file created by Marc Hausmann
  - Production target: C @ 10 mm
  - Wedge-1: Al @ 2.5 mm
  - Wedge-2: Al @ 3.4 mm
- Only changed Brho & Power
  - For 8H, we reduce primary beam energy from 250 to 140 MeV/u.
- Adopted 50% transmission to SOL
- Adopted Diff-Eff efficiency of 0.5%

Beam	Half-Life	Gamma Ray Energy [MeV]	Detection Efficiency	Count Rate (Implantation)	Expected Catcher Time (3% stats)
^{35}Ar	1.78 s	1.22 MeV	5%	600 cps	6 min
^{34}Ar	846 ms	0.67 MeV	2.5%	540 cps	7 min
^{32}Cl	300 ms	2.23 MeV	3%	2076 cps	2 min
^{31}S	2.5 s	1.27 MeV	5%	32 cps	2 h
^{16}N	7.3 s	6.13 MeV	0.01%	0.3 cps	177 h
^{15}O	122 s	0.51 MeV	12%	1698 cps	2 min
^{14}O	71 s	2.31 MeV	3%	18 cps	3.5 h
^{13}N	10 min	0.51 MeV	12%	1061 cps	3.5 min
^{8}He	120 ms	0.98 MeV	6%	2 cps	27 h

Element Dependence Release
Mass Dependence can be Neglected in First Order

- Release properties can be studied by studying the release efficiency as function of the temperature
  - From the release curve, Arrhenius coefficients for diffusion can be obtained
- FRIB/ARIS allows studying simultaneously the release for several elements at the same time, by using cocktail beams or fast changing magnetic rigidity of ARIS
- Elements which could be studied in one run (about 3-4 days) for one stopper
  - He, N, O, S, Cl, Ar and Kr
  - Stopper choices to be decided

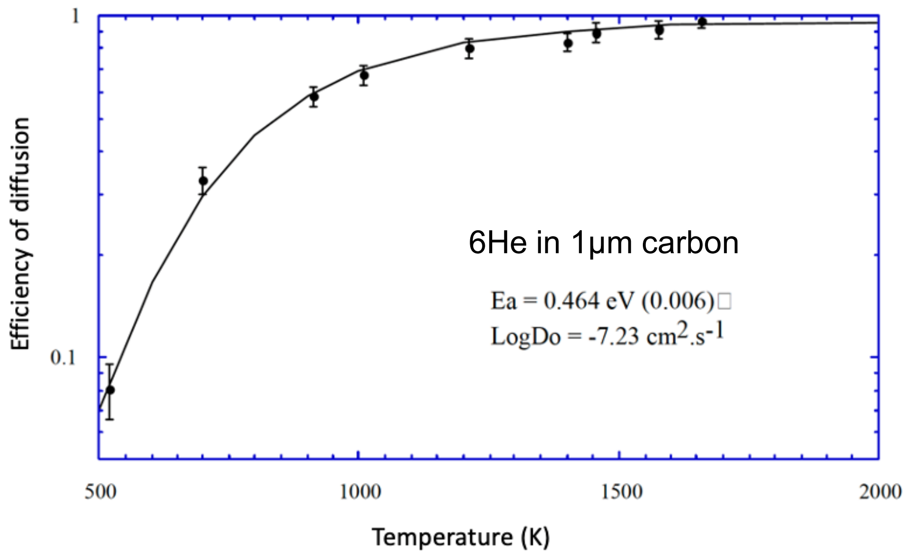
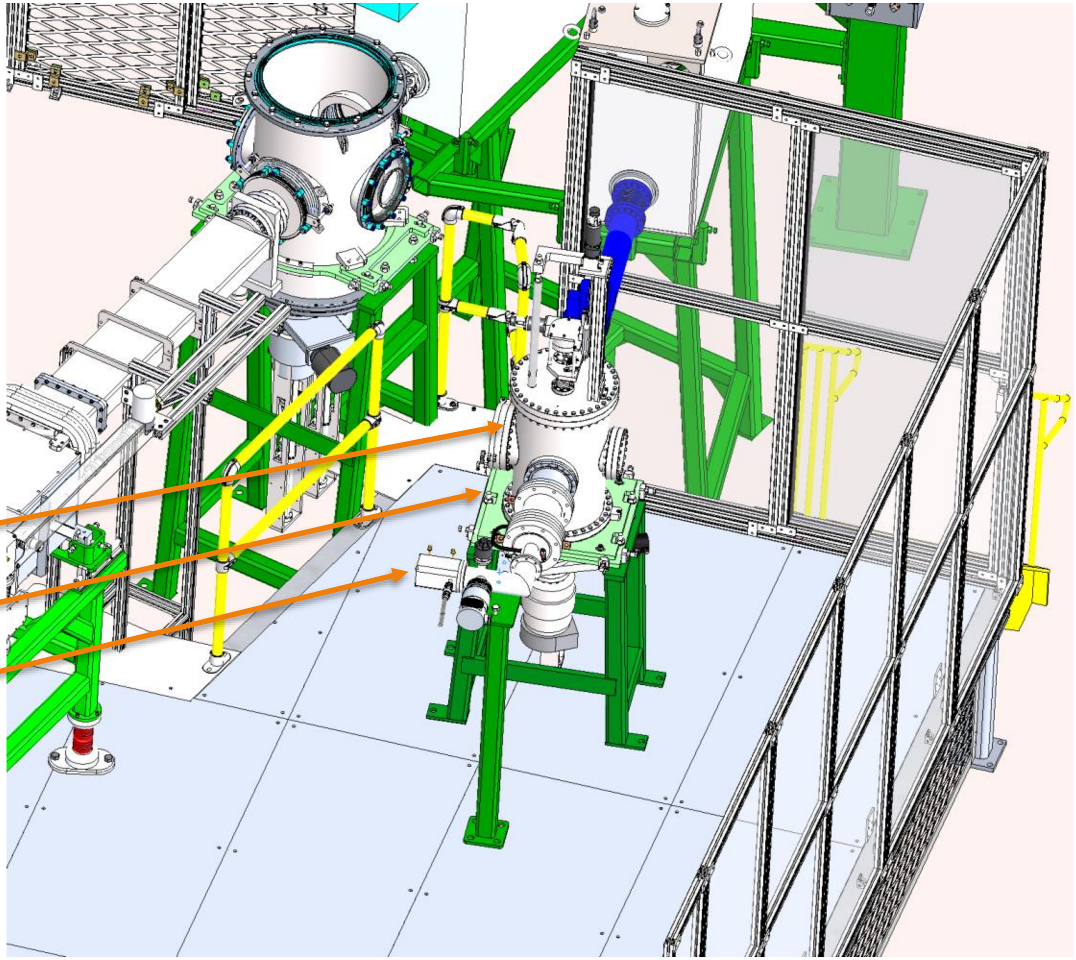


Figure adapted from: Pellemoine, Thesis 2001 https://hal.science/tel-00008873v1

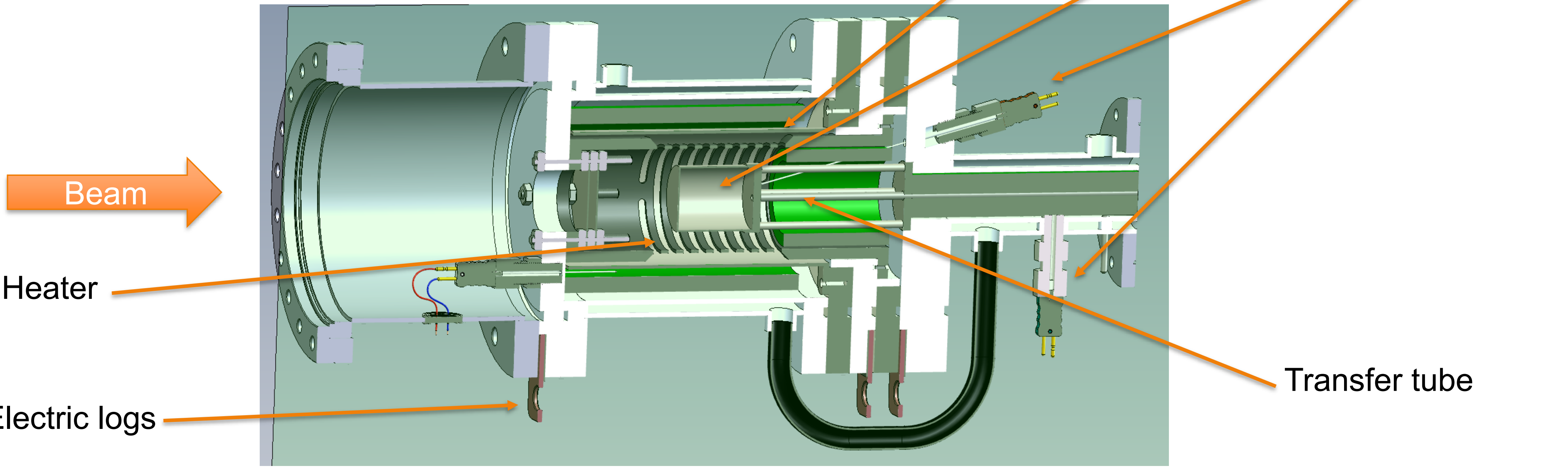
SOL to be Installed in N4 Vault

- The full SOL compact system will be installed in N4 vault between ACGS and RTGC
- HE beamline elements in place
- Degrader box (with degrader system) recovered
- Scintillator detector being developed
- Stopping chamber being designed



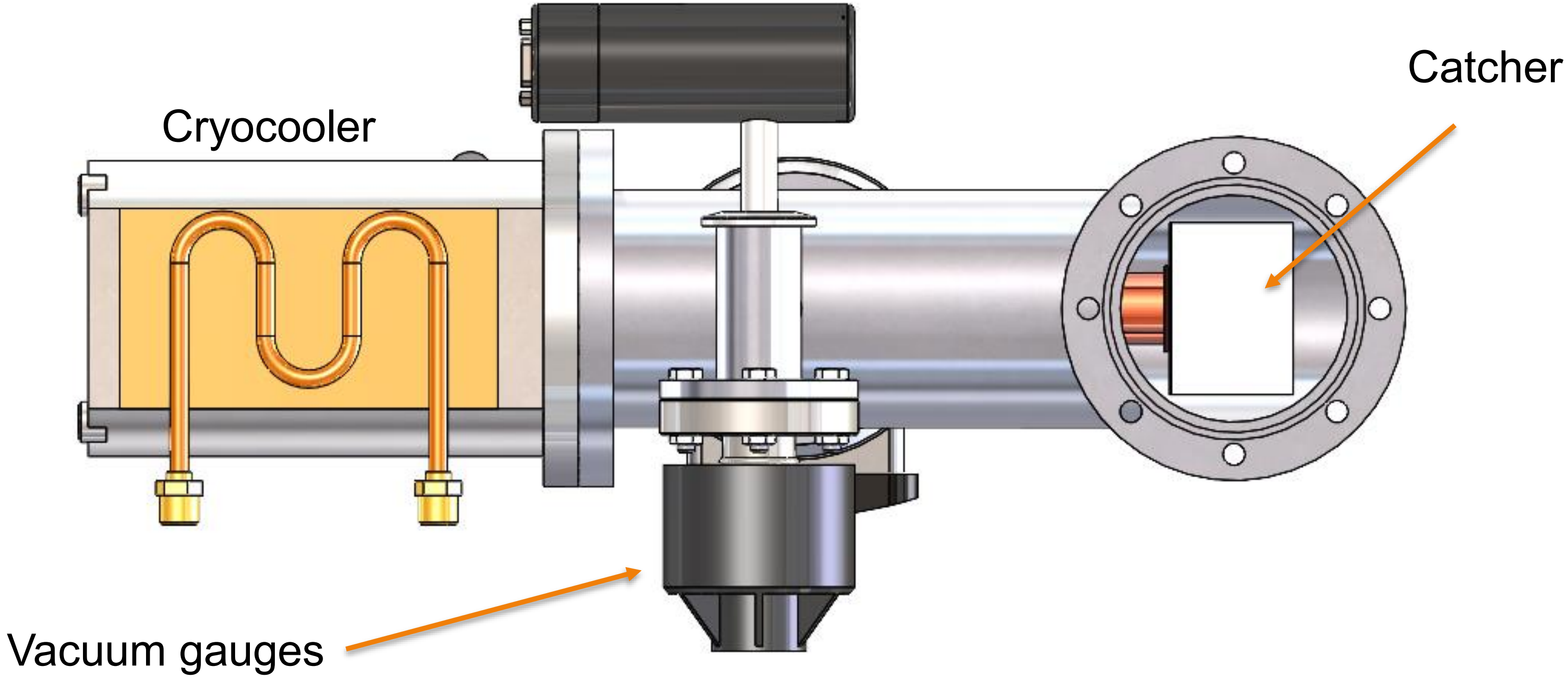
Design of the Stopper System Well Advanced

- Design of the stopper chamber is well advanced
  - Most elements are of carbon, some coated by diamond-like-carbon
  - Carbon insulation is done by Papyex or graphite felt
  - Electric insulation done by alumina flanges
    - » Flanges are water-cooled, not shown



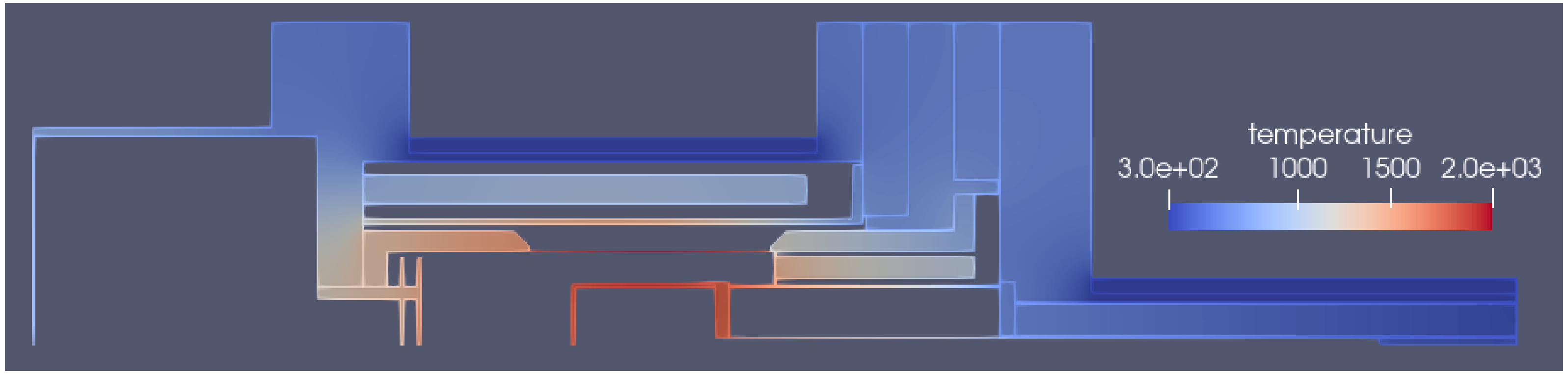
Design of the Cryo-catcher Well Advanced

- The catcher uses a cryo-cooler – same model of ACGS – PT30 pulse tube from Cryomech
  - Provides 37W at 80K, enough to collect effused isotopes out of transfer tube
- Catcher is on copper, placed in front of the transfer tube

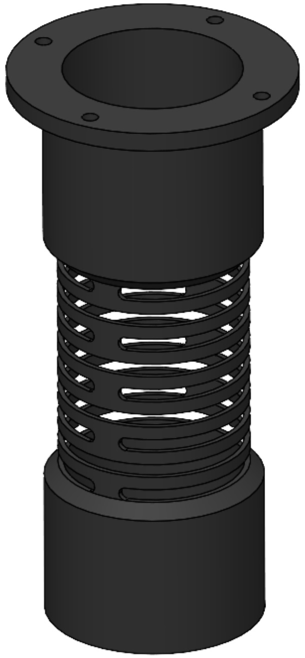


Thermal Studies of Stopper Chamber Status

- Temperature distribution is under study
  - Corners of the system are still too hot
  - New design involving cooling of the flanger should improve flange temperatures
  - Design of the system-temperature calculations are being done interactively
- Total power to heat the stopper (today's design) is 4.3 kW
- Total power to heat the transfer tube (today's design) is 1.2 kW

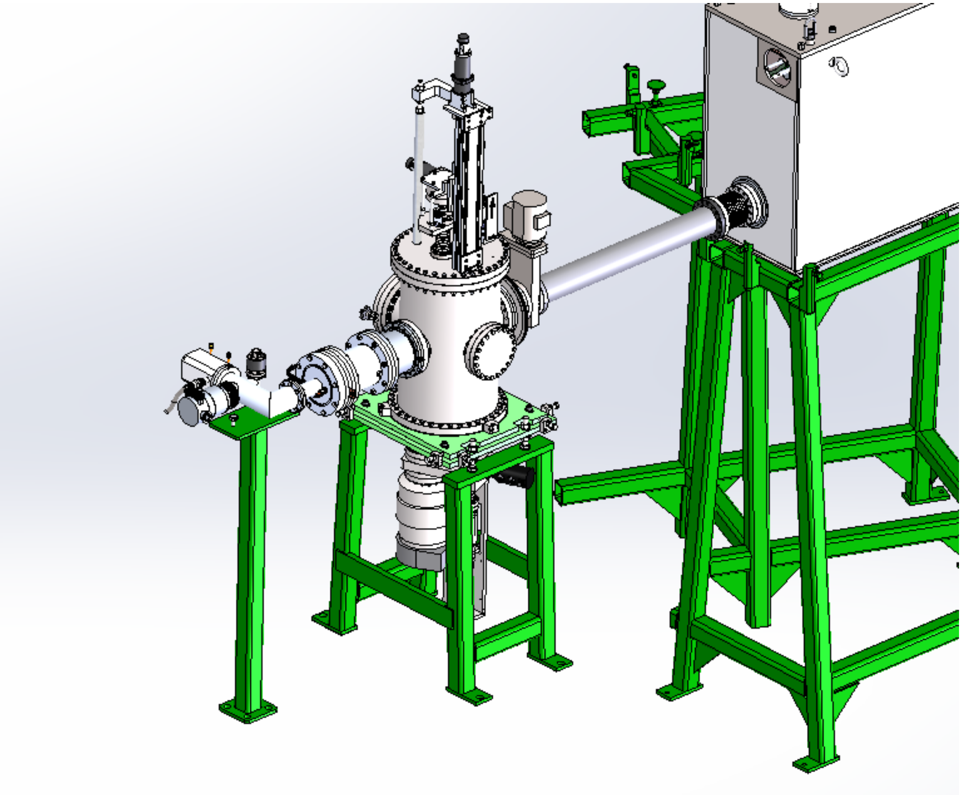


Heater



SOL Installation in N4 For 2026 Summer Shut-Down

- Note: start of SOL suffered from requests to perform a hazard analysis and
- Schedule review for SOL
  - 30% review in December 2025
  - Finalize design of all components by February 2026 (90% review)
  - Assembly and installation in N4 vault by September 2026
  - Tests off beam by November 2026
    - » Temperature validation of components
    - » Vacuum and out-gassing
    - » Choice of stopping materials
  - Measurements with beam starting in January 2027
  - End of the project forecast to April 2027
    - » Expect 6 months request for no cost extension from initial planning



Original Estimated Timetable

Task Objective 1	Q 1	Q 2	Q 3	Q 4	Q 5	Q 6	Q 7	Q 8
Simulation of effusion from ion stopper and transport to cryogenic catcher	■							
Evaluation of heating concepts for the container oven	■	■						
Mechanical design of the SOL Setup		■	■					
Procurement of cryocooler and other components			■	■				
Fabrication of the SOL setup including the container oven			■	■				
Installation of the existing wedge and diagnostic box at beam line and the SOL setup				■				
Test of the SOL setup without beam				■				
Task Objective 2	**Q 1**	**Q 2**	**Q 3**	**Q 4**	**Q 5**	**Q 6**	**Q 7**	**Q 8**
Release tests with noble gases					■	■	■	■
Release test of oxygen beams in form for CO or CO2						■	■	■
Release tests for non-gaseous elements using NF3 and other materials						■	■	■
Documentation of release results					■	■	■	■

Budget Status

- Note
  - Due to priority, personnel allocation in this first phase (conception design), consisted of 2 PIs and 2 undergraduate student
    - » Molly Jesper for design work
    - » Guilherme Roda for diffusion-effusion simulations
    - » Design finalization and parts purchasing are moving to FRIN engineers – not included in first year the expenses

	Year 1 ($)	Year 2
a) Funds allocated	485,000	300,000
b) Actual cost to date	59,982	0

Summary

- FRIB provides fast, stopped and reaccelerated rare isotope beams for science
- Technique of stopped beams is well mastered at FRIB
  - Gas stoppers are the first choice, however limitation on beam intensity and light ions need to be addressed
  - Solid stoppers can unlock solutions for selected isotopes
  - FRIB opens opportunities to new material development in particular nanomaterials, which can present fast release
- SOL - Funding opportunity DE-FOA-0003261 granted
  - SOL design is well advanced
  - Schedule is defined
- SOL will request a non cost extension of 6 months