Reaction Studies on Unstable Nuclei at NIF

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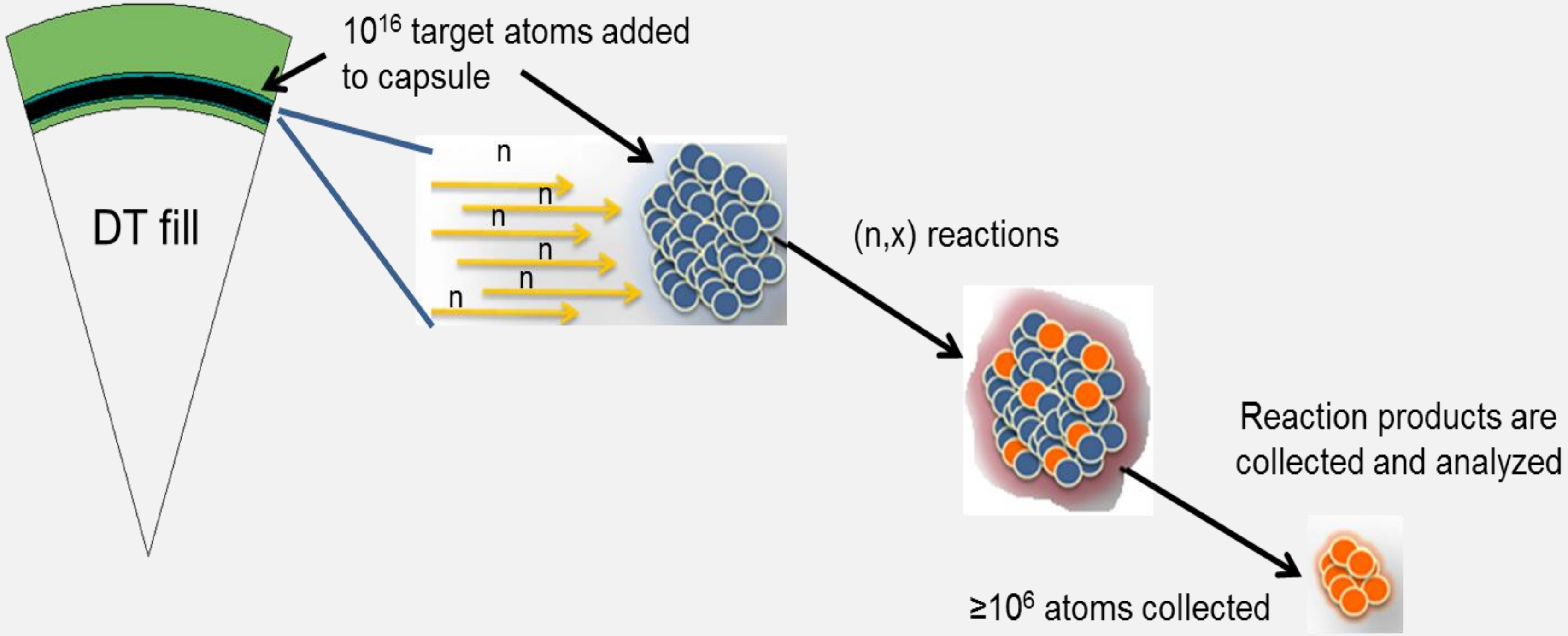
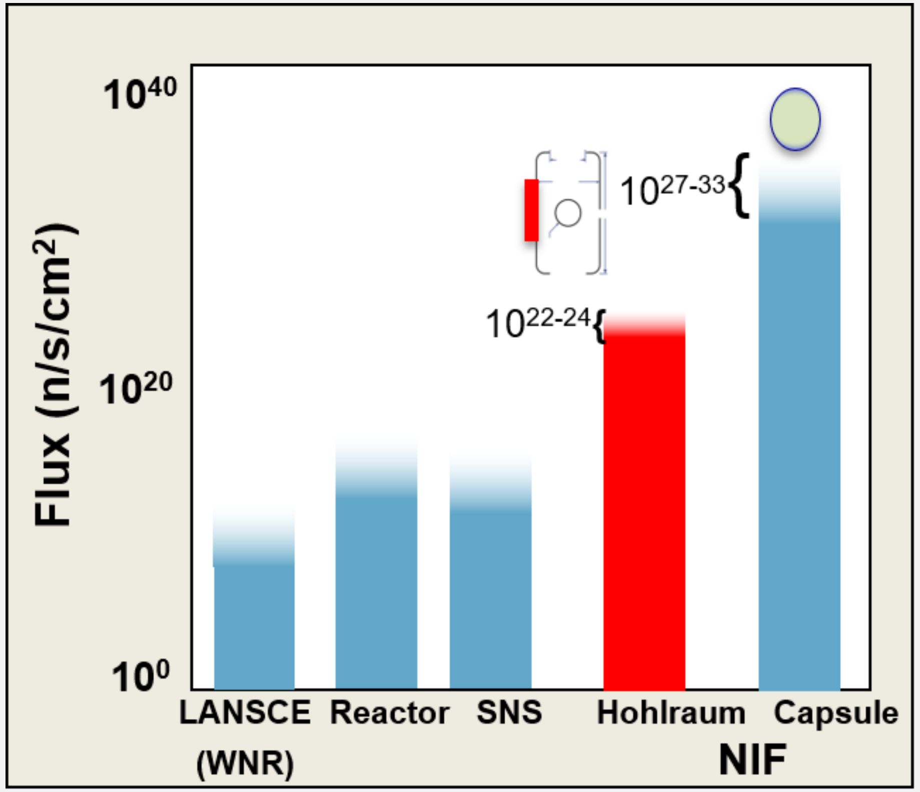
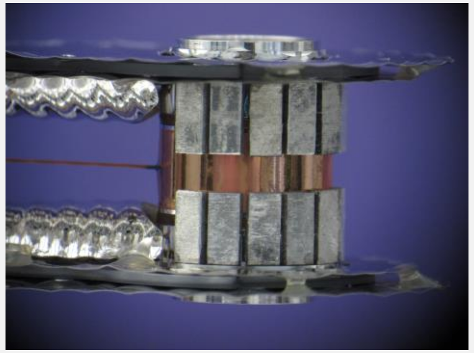
NIF is a unique tool to study neutron-induced nuclear reactions

Target material added to capsule

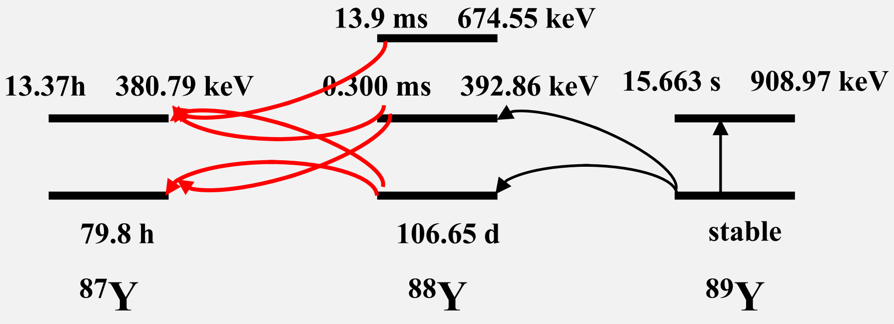


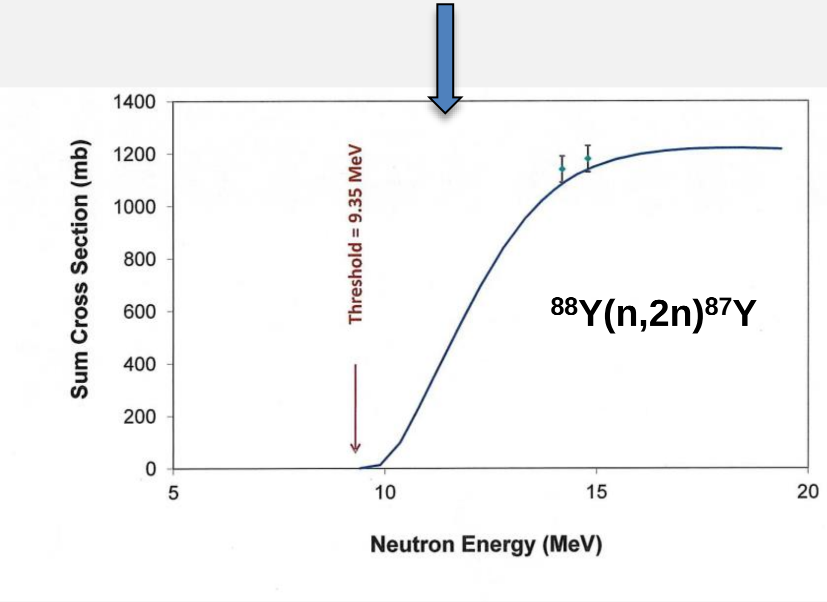


Target material added to hohlraum



Program goal: Develop doped capsule platforms for measuring radiochemical tracer nuclear reactions for Stockpile Stewardship





Only 2 measurements exist for this reaction

- Thermonuclear performance was determined through radiochemical detectors
- Reactions on short-lived, excited states must be included in performance models, but cannot be measured directly
- NIF-based measurement requires ~10^{13} atoms (ng, µCi) of a radioactive target compared to ~>10^{18} atoms (~mg, ~>1 Ci) for an accelerator target due to increased neutron flux
- Current: perform a series of three NIF shots to measure the ^{89}Y(n,2n)^{88}Y, ^{88}Y(n,2n)^{87}Y and determine if Y fractionates from other rare earth elements during a NIF shot

Yttrium will be done first for method validation; other detector elements also need cross-section data

Reference reactions are used to determine unknown cross sections

- The product of the reaction of an "instantaneous" pulse of neutrons with a detector element (n^0) is given by:

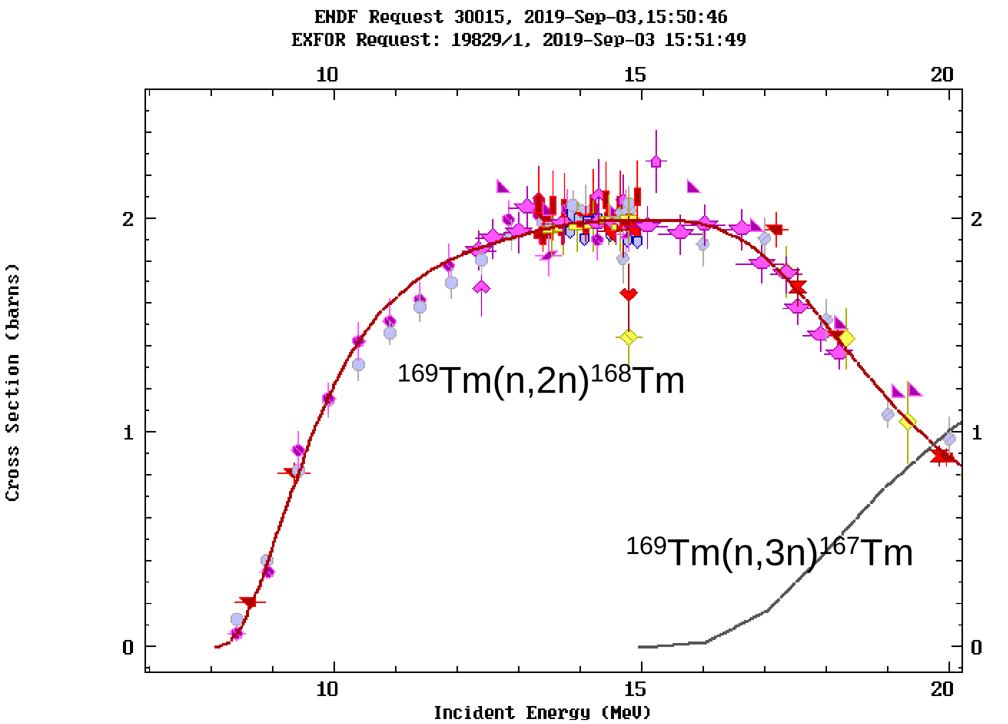
$$N = n^0 \phi \sigma$$

- If two target nuclides are co-loaded together, one with a known cross section (σ), the flux (ϕ) cancels:

$$\sigma_{unknown} = \sigma_{known} \, (n^0_{known}/n^0_{unknown})(N_{unknown}/N_{known})$$

- Uncertainty is related to errors in the known cross section, loaded target assay, counting statistics, and some correction factors (e.g., differing reaction thresholds)

- ^{169}Tm(n,2n) is the "known" reaction for ^{88}Y(n,2n)



Uncertainty on measured first-order (n,2n) at NIF will be ~5% depending on nuclide

Radiochemistry diagnostics at NIF have expanded

Solid debris collection

Vast Area Detector for Experimental Radiochemistry (VADER), ~1%

Solid debris collection

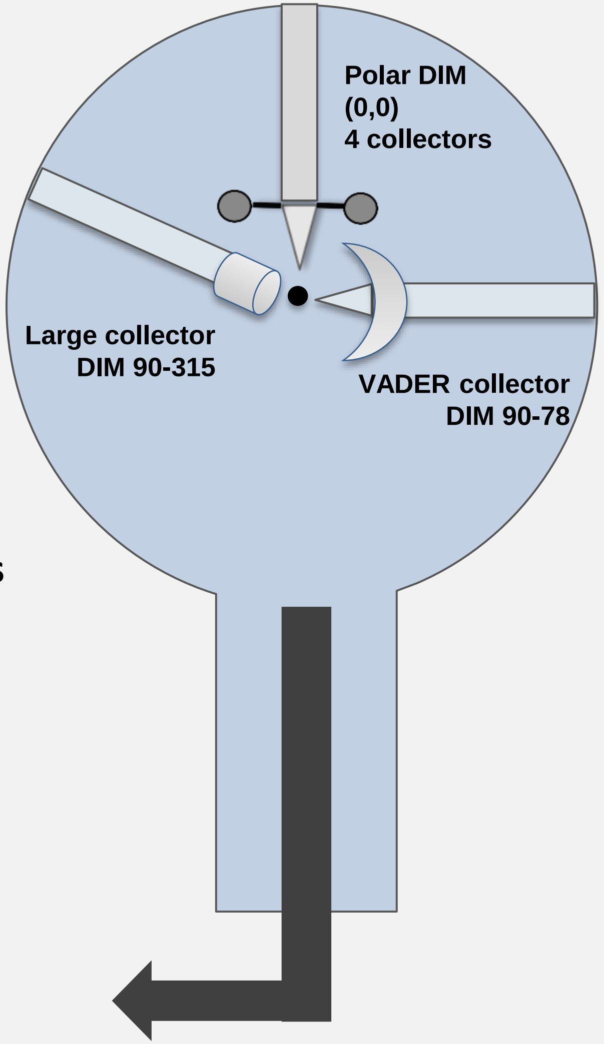
Large area solid radiochemistry collector, 1-5%

Solid debris collection

Solid radiochemistry collectors (4), ~0.1%

Collection of noble gas products

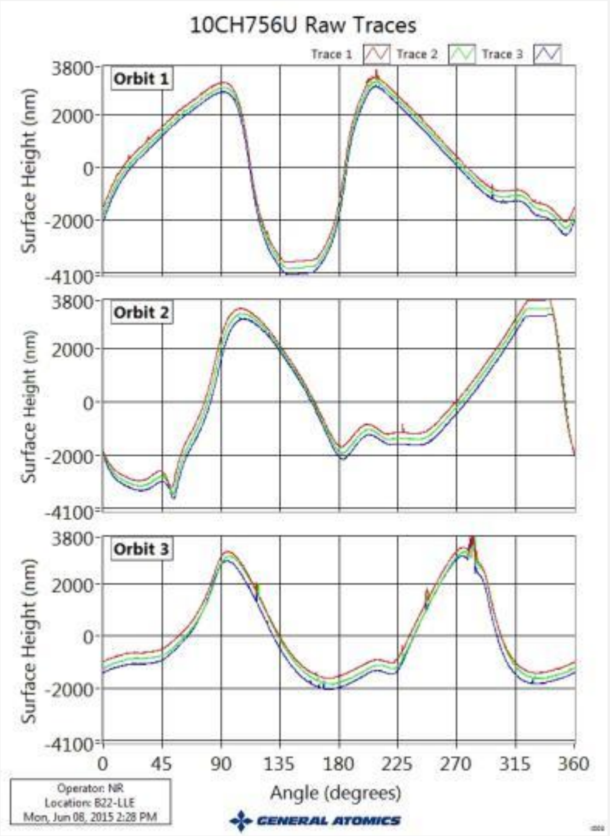
Radiochemical Analysis of Gaseous Species (RAGS)

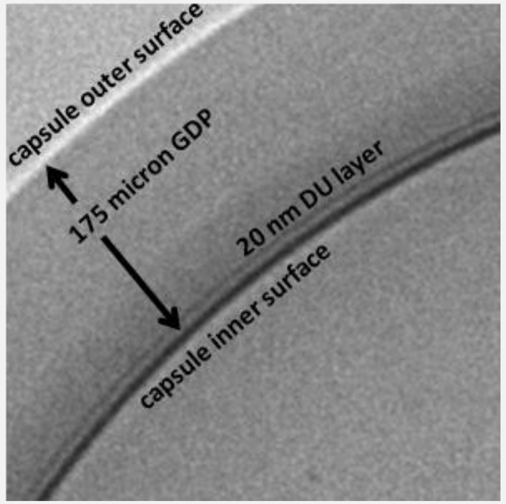


Neutron yield, neutron imaging, bang time, x-ray imaging, ion temperature are provided as standard NIF diagnostics

^{238}U capsules showed the need for alternate doping methods

- DU sputtered in the ablator shell causes outer surface deformations
- The time required to produce a full capsule is not amenable to using radioactive species





DU layer in CH capsule (GA)

Capsules with dopants on the inner surface had consistent neutron yields

	N181104-001	N190103-001	N190519-001
scale	sub	sub	sub
laser	SScAS_2DConA	SScAS_2DConA	SScAS_2DConA
laser energy	0.8 MJ	0.9 MJ	0.85 MJ
hohlraum	469 Au tamped	469 Au tamped	469 Au (tamped)
Dopant	**^{7}Be**	**^{238}U**	^{7}Be
Dopant layer recess	0 µm	0 µm	0 µm
Y_{DT}	1.5 x 10^{14}	3.2 x 10^{14}	5.3 x 10^{14}
Tion (keV)	2.56	2.61	2.82
DSR (%)	**0.5**	0.9	0.85
Fill tube	30 µm	10 µm	30 µm
Doping method	Injection	Vacuum	Injection

Simplified Flow Diagram for Capsule Doping

Years before shot

Development of radionuclide cocktail for desired measurement

→

Up to a year prior to shot

Production of radionuclides required for doping

→

Finish 4 months prior to shot

Purification of radionuclides and production of doping cocktail…Requires new chemical methods (for every different dopant) to produce ultra-clean samples

4 months prior to shot (takes ~1 month)

Receive drilled capsules from GA and Doping of NIF capsule and characterization of what is added

→

3 months prior to shot

Capsules Sent back to GA for CFTA assembly

→

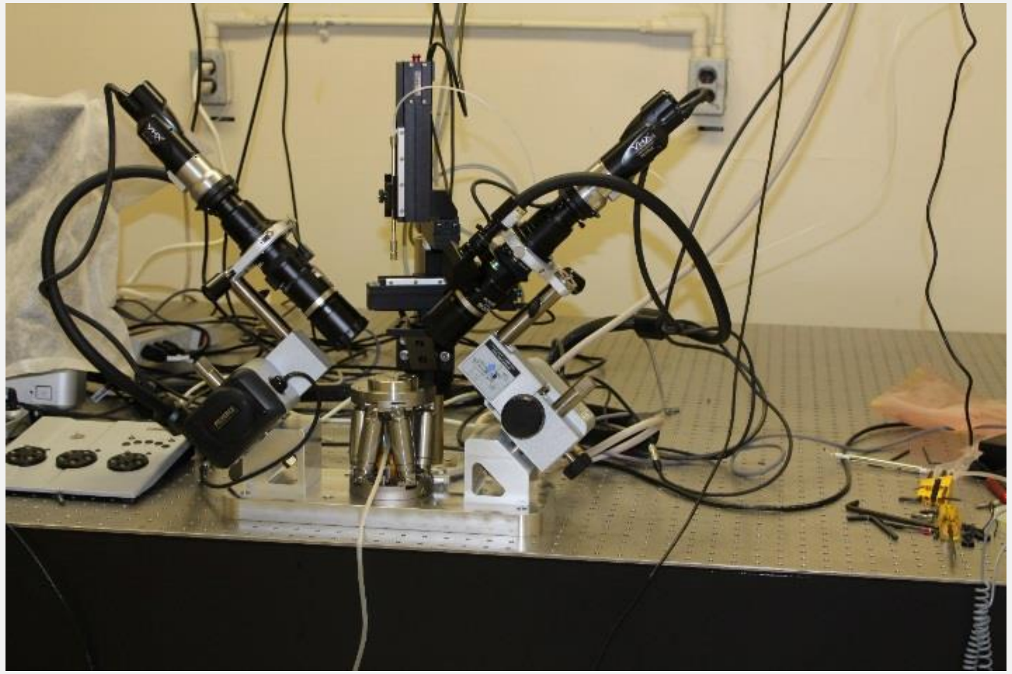
2 months prior to shot

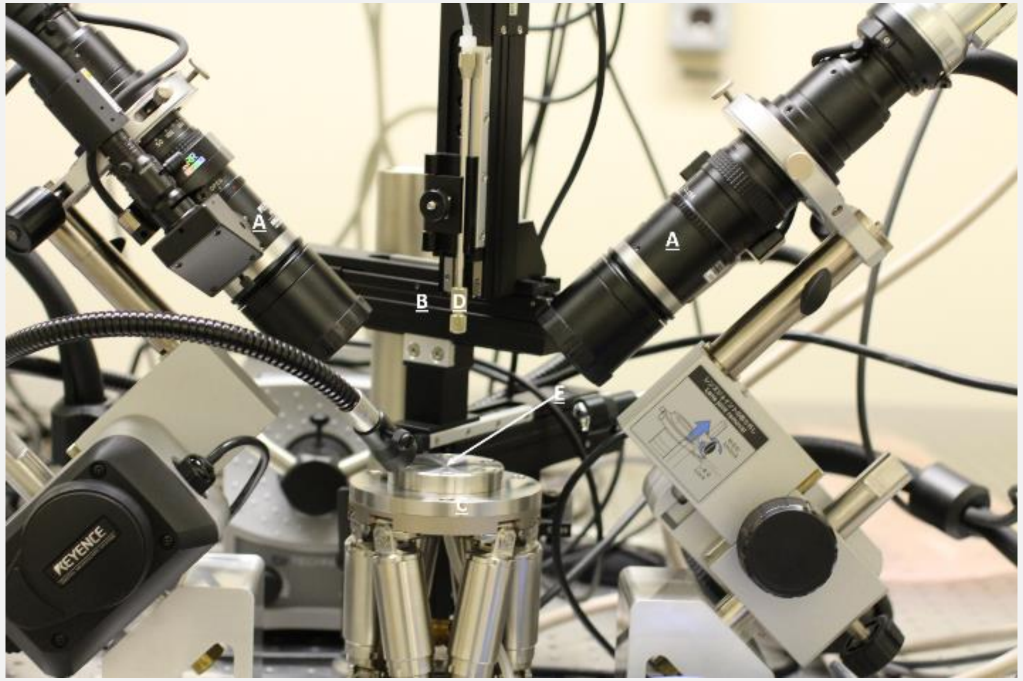
Capsules to LLNL for target assembly

↓

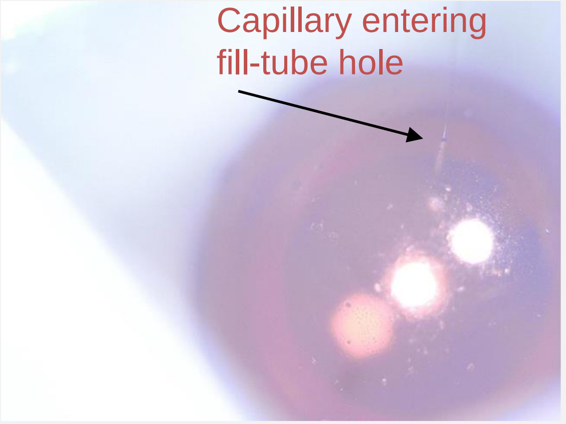
SHOT

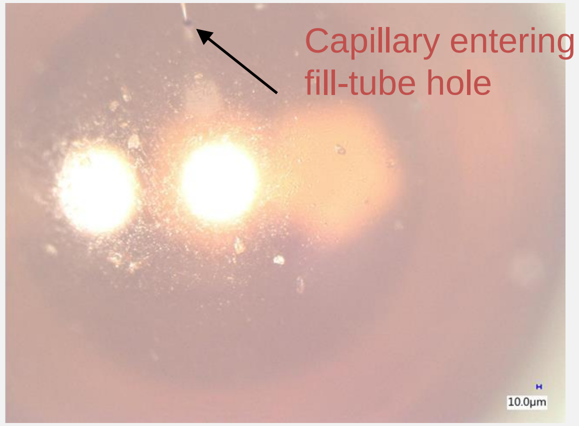
Apparatus for NIF Doping: Automated Robotic Injection System for Targets (ANDARIST)





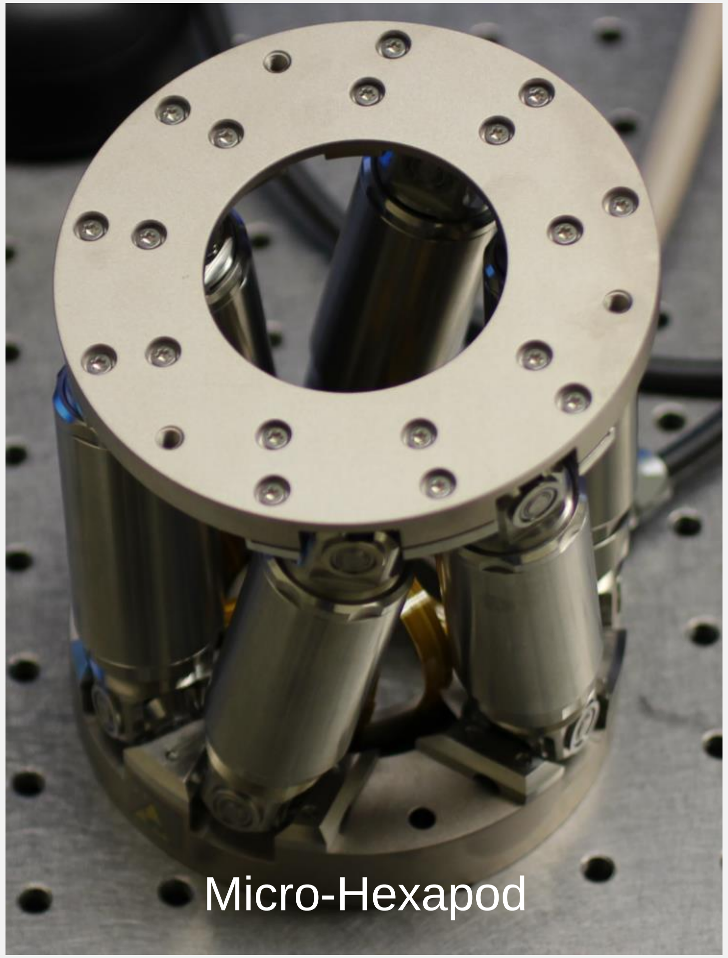
- System allows for precision alignment of microcapillary with fill-tube hole for injection
- Each material "cocktail" requires R&D to optimize the matrix and mass loadings

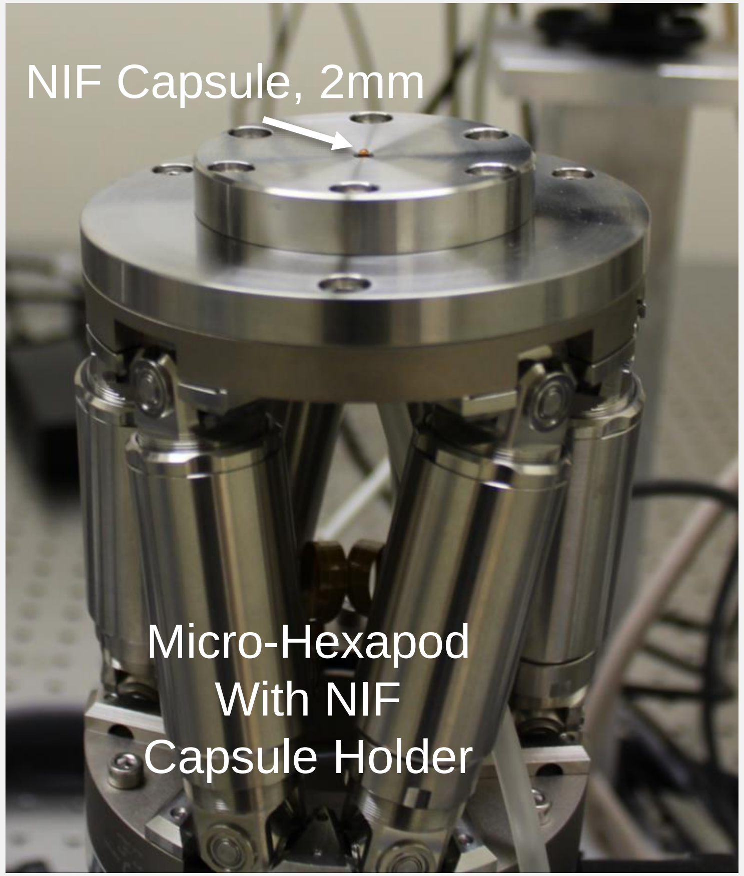




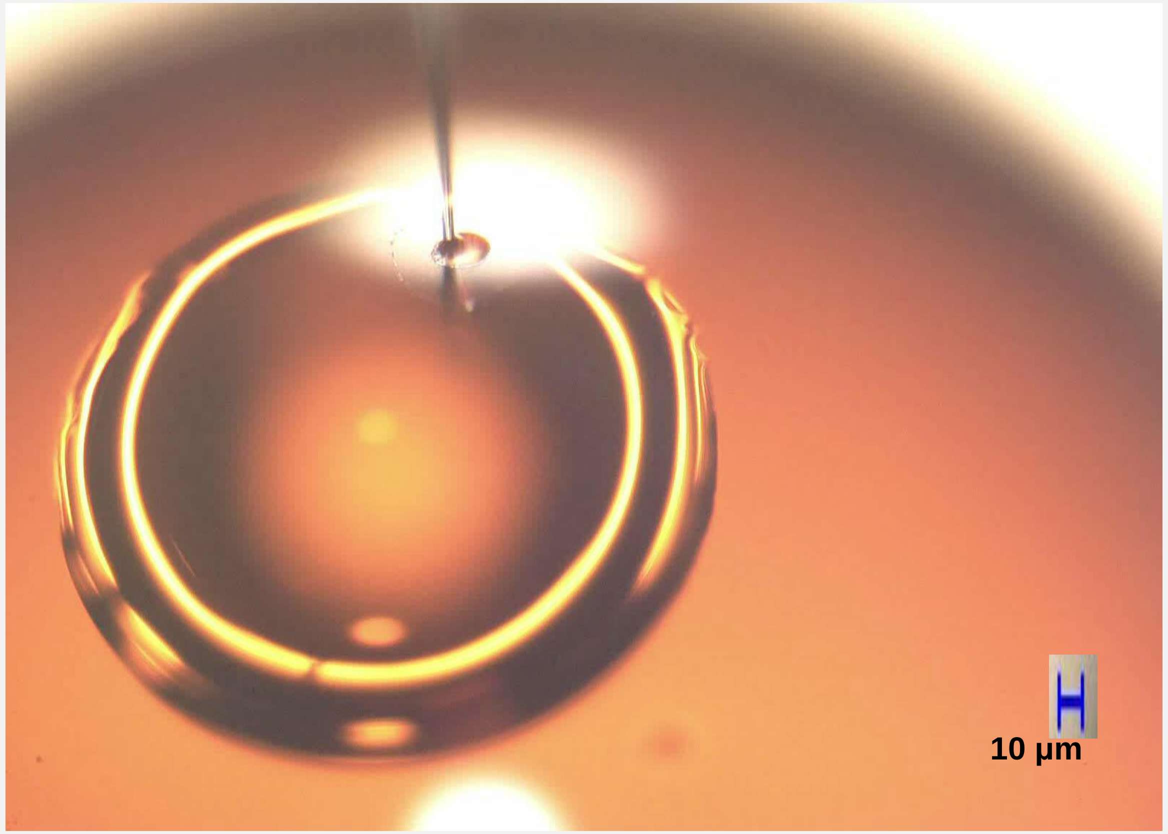
First doped capsules contained ^{238}U and $^{7,10}Be$ (^{7}Be $t_{1/2}$=53 d)

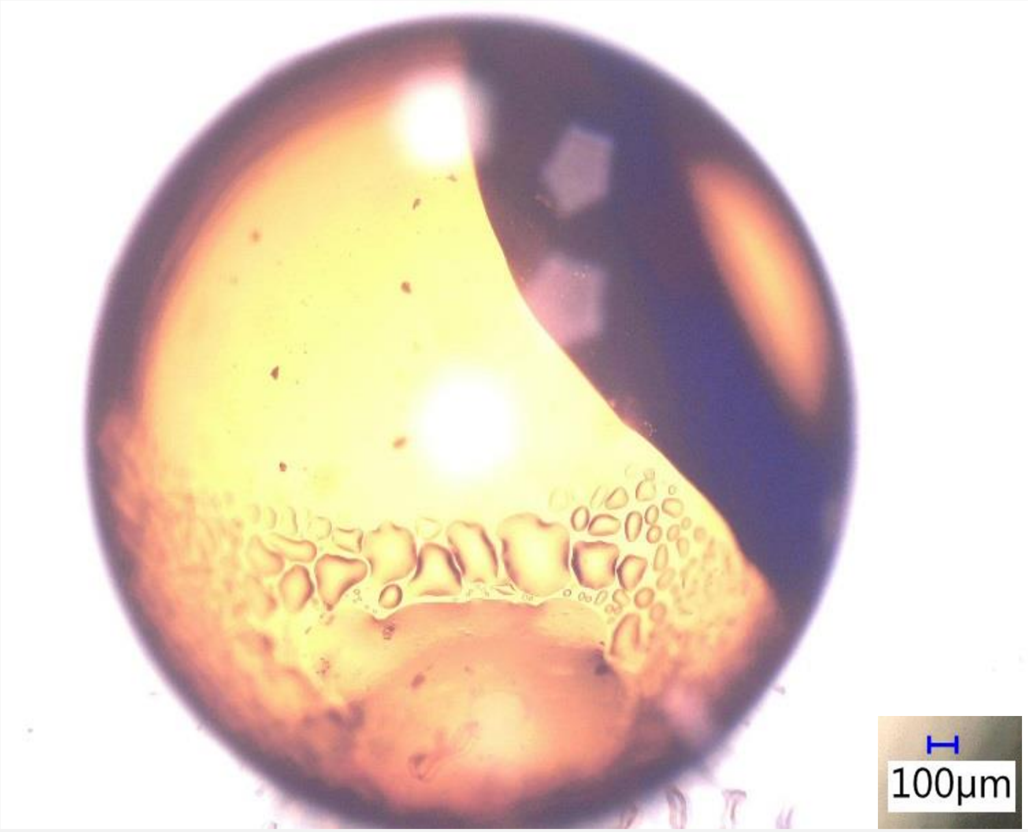
Heart of ANDARIST



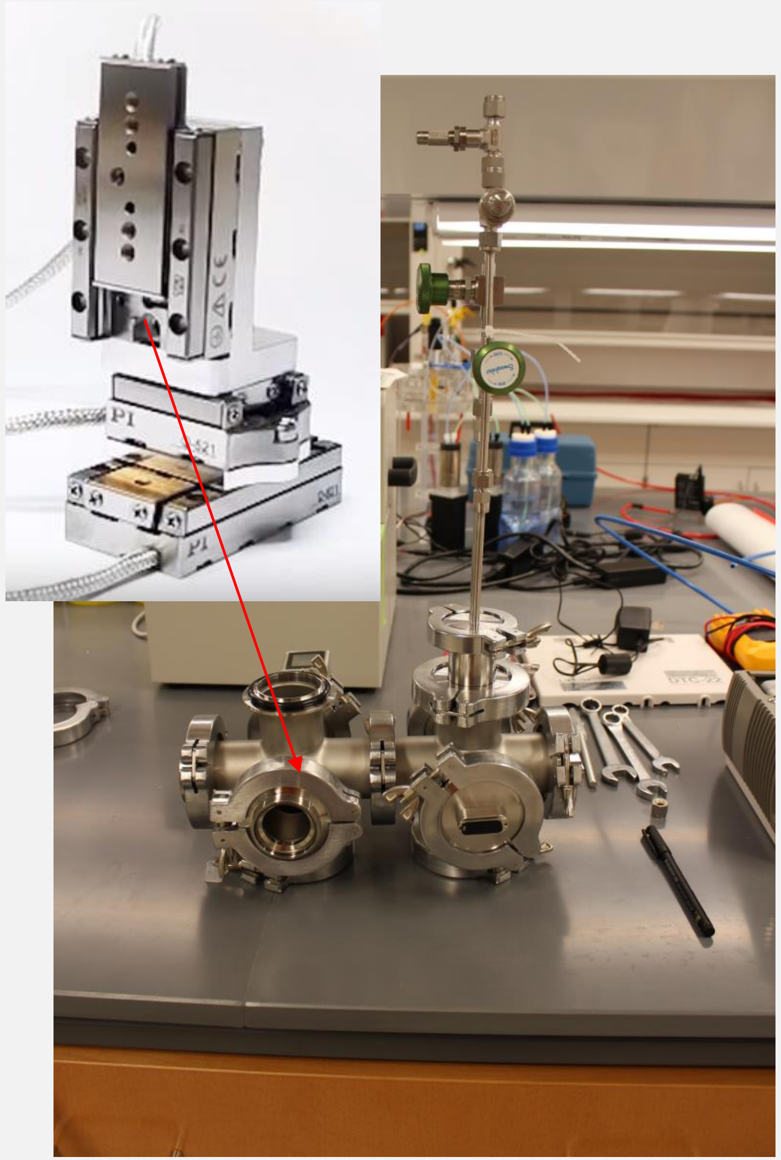


Microinjected NIF Capsule





VORCAN: Vacuum Optimized Radionuclide-to-Capsule Administer for NIF



- Improved vacuum filling system for robust addition of radionuclides to NIF capsules, in volumes slightly larger than ANDARIST
- Very small XYZ manipulator in vacuum chamber, capable of 0.4 nm movements
- Allow submersion of capsule in <4 μL of liquid
- Filled by pressure difference
- Simpler and quicker method for addition of dopants to capsule, when larger volumes are available

Comparison of Systems

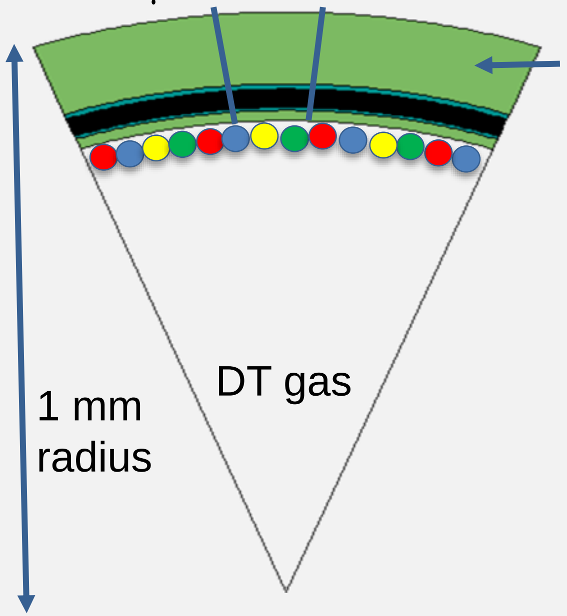
ANDARIST

- Very small solution volumes possible (< 1 μL)
  - Low production radionuclides possible
- Challenging operation, ultra clean samples required.
  - Need new chemical methods for sample production for each radionuclide
- Any different solution requires different injection parameters
- Minimal touching of outside of capsule potential to keep it clean

VORCAN

- Small solution volumes possible (~3 μL)
  - Some low production radionuclides possible
- Easy operation
  - Solution cleanliness not as important
- All solutions are injected the same way, by pressure difference
- More touching to the outside of capsule, potential for damage if not careful
  - Not a big concern for our program

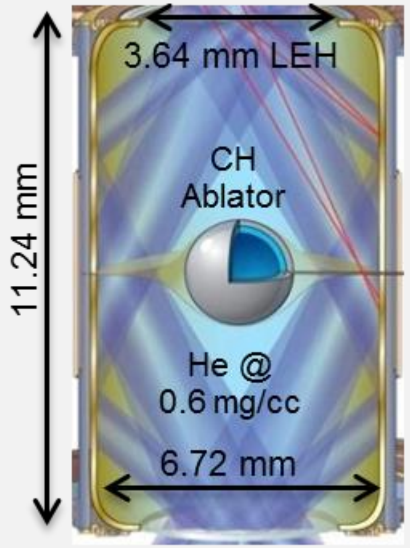
Proposed capsule for the first ^{89}Y shot has a complex mixture of tracers



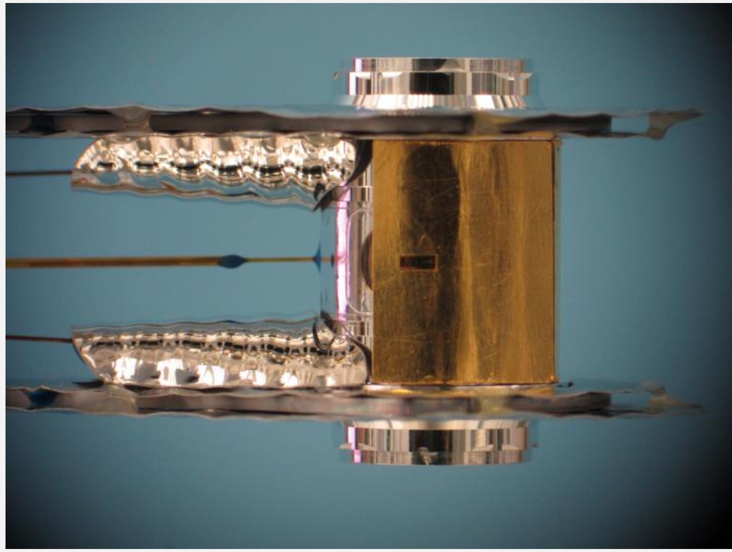
Nuclides deposited on inner surface:

^{89}Y (stable, ~10^{14} atoms) – "unknown"
^{169}Tm (stable, ~10^{14} atoms) – "known"
^{91}Y ($t_{1/2}$=59 d, ~μCi) – Y collection tracer
^{152}Eu ($t_{1/2}$=13 y, ~μCi) – Tm collection tracer

Gold hohlraum (no DU present)



Final target assembly – hohlraum wrapped with extra gold to enhance debris collection



Production of ^{91}Y, target doping and shot date have been coordinated to account for the half-life

This and two subsequent capsules failed cryogenic pressure tests at NIF, radiation damage to plastic shell

Previous Capsules

CH Capsules

- Had arrived at GA (previous couple shots) with clear fill holes on the vast majority of capsules
- These capsules are transparent, our optical microscopes were rather good at seeing salt in the fill holes/addressing this issue before sending

HDC Capsules

- Optically non-transparent, so ability with current set up for CH to see through fill hole was limited.
- Resulted in capsules that were plugged to a degree much more severe than CH capsules
- New, QA/QC procedure being developed to address this

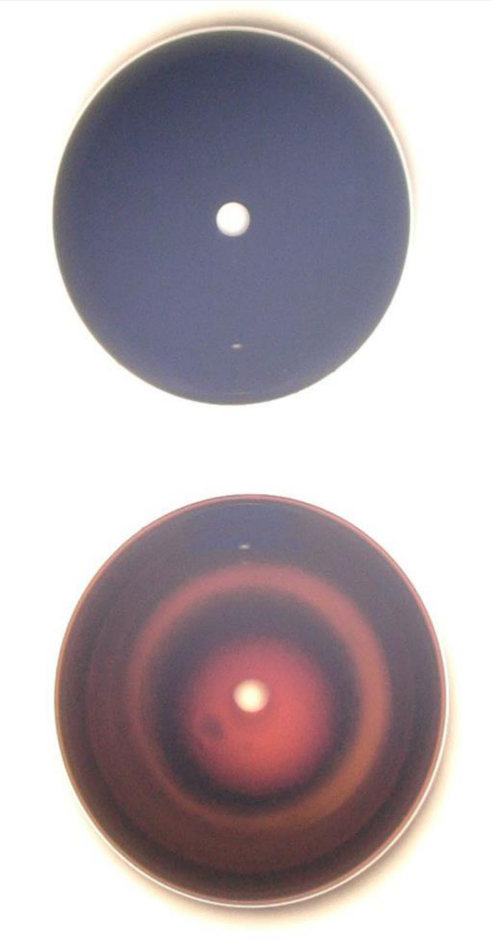
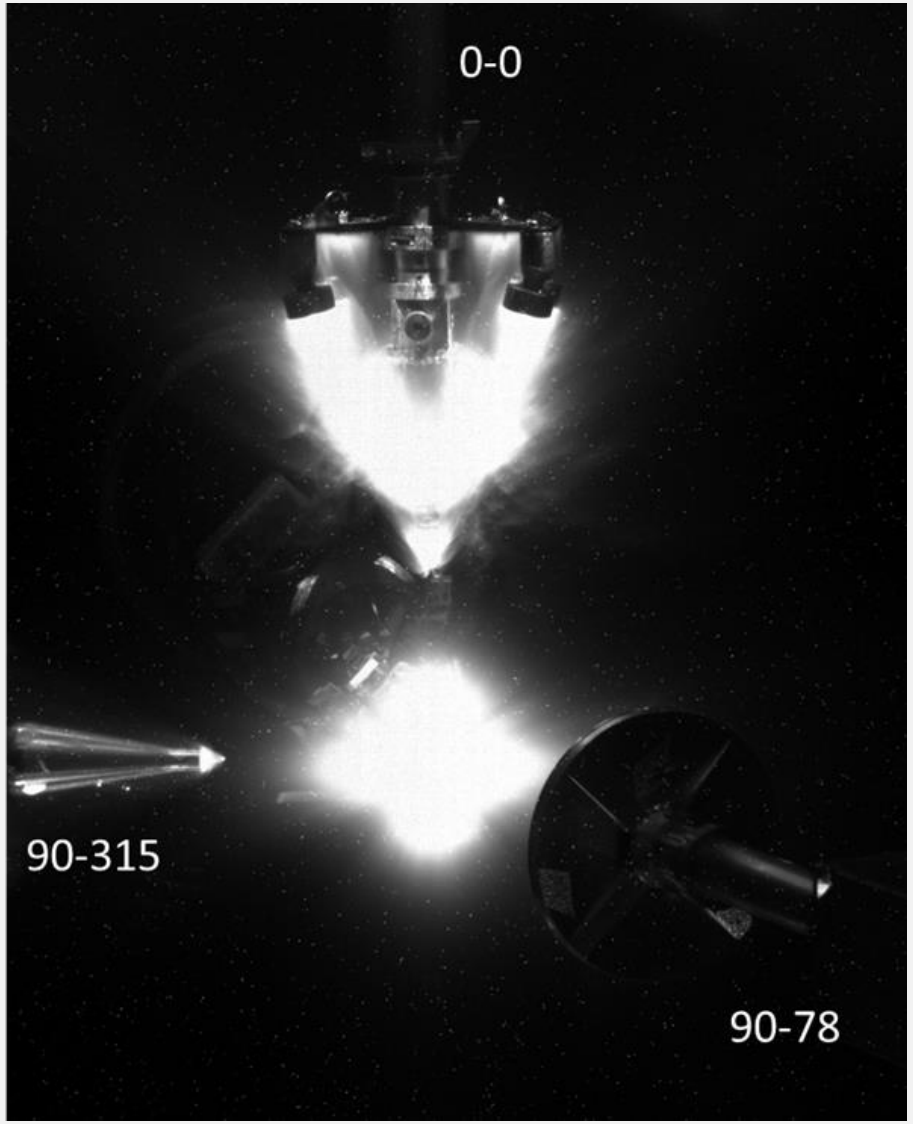


Figure: HDC (top), CH (bottom)

Next steps for radiochemical measurements at NIF

- Current shot request (requesting this March for FY23):
  - ^{89}Y capsule with improved doping method for HDC ($^{89}Y(n,2n)^{88}Y$)
  - ^{88}Y capsule ($^{88}Y(n,2n)^{87}Y$)
  - Fractionation capsule with mixed radioactive rare earth elements to ensure rare earths are collected with the same yields
- Future: apply methods to other unknowns such as $^{168}Tm(n,2n)$
- Future: evaluate possible (n,γ) reactions on radioactive species (DD shots)



Conclusions

- Doping of NIF capsules is a long process that requires coordination between radiochemists, GA and target fab to work on complex timelines
- The production and use of the radionuclide cocktails for doping requires new chemical methods developed for every single dopant used
- A microinjection doping method, ANDARIST, has been developed and tested for use with rare radionuclide cocktails on small volume scales
- An improved vacuum based doping system, VORCAN, is being brough online for robust addition of radionuclides to NIF capsules, at a slightly larger volume scale than ANDARIST (which will make doping of most things more straight forward)
- HDC impose a unique challenge and aid in the radiation resistance of the targets

Acknowledgments

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