SHINE Chemistry Overview

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Argonne National Laboratory

2015 Mo-99 Topical Meeting

Boston, MA

August 31 – September 3, 2015

SHINE Medical Technologies

- SHINE Medical Technologies is dedicated to being the world leader in safe, clean, affordable production of medical tracers and cancer-treatment elements
- SMT and its partners have developed a system that can produce reactor-grade medical isotopes without a nuclear reactor
- Technology has two key aspects
  - Primary neutrons created by high-output D-T source
  - Neutrons enter an LEU solution where they multiply sub-critically and create medical isotopes
- Initial construction will produce nationally relevant quantities of Mo-99 and other medical isotopes (50% of U.S. Mo-99 demand)

Argonne's Role in Supporting SHINE

- Major tasks
  - Preparation of the uranyl sulfate target solution
  - Development and design of the Mo-recovery system using TiO_2 sorbent
  - Use of the LEU-Modified Cintichem process for Mo purification
  - Periodic cleanup of irradiated target solution
  - Radiation stability of system components and peroxide formation using the Van de Graaff
  - Developing an understanding of radiolysis effects on
    - Solution chemistry
    - Gas generation
    - Precipitation
  - **Mini-SHINE experiments**
  - Micro-SHINE experiments

Mini-SHINE Experiments

- Argonne's mini-SHINE experiment will irradiate aqueous uranyl-sulfate solutions using an electron linac to:
  - Study the effects of fission on target-solution chemistry and radiolytic off-gas generation
  - Demonstrate the recovery and purification of ^{99}Mo from an irradiated target solution
  - With the assistance of PNNL, sample off gas for Xe, Kr, and I
  - Ship Mo-99 product to potential Tc-99m generator manufacturer partners

Phase 1

- Linac will be operated initially at 35 MeV and 10 kW beam power on the target
- 5 L solution will be irradiated with neutrons generated through gamma-n reaction in tantalum target
- Maximum solution power will be $\leq$ 0.05 kW/L
- Up to 2 Ci of Mo-99 will be produced

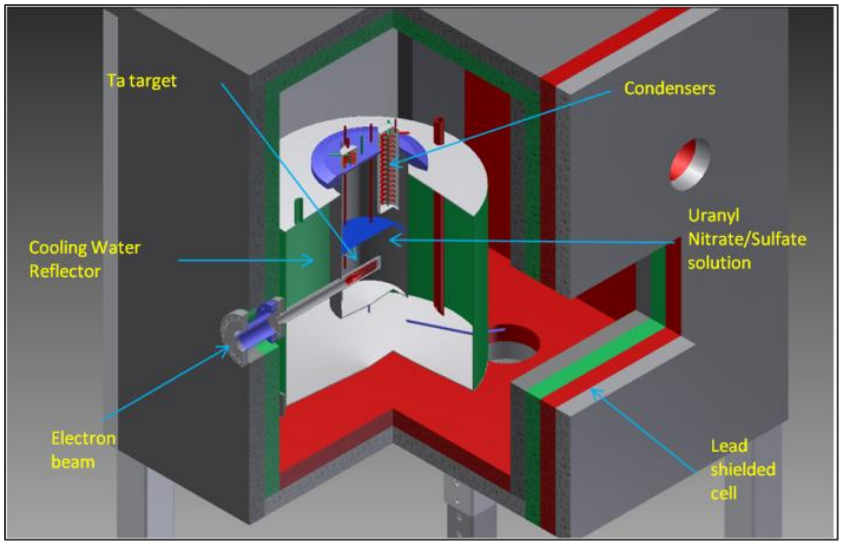
Phase 2

- Experiment will be conducted at 35 MeV beam energy and up to 30 kW beam power
- 20 L solution will be irradiated with neutrons generated in a depleted-uranium (DU) target
- Maximum solution power will be $\leq$ 0.5 kW/L
- Up to 20 Ci of Mo-99 will be produced

Mini-SHINE Progress

- Phase-1
  - Conservative approach
    - H_2O and $NaHSO_4$ tested first
    - To verify all system components before producing fission products
  - Radiation stability of components were verified using a Van de Graaff generator
  - Water and sodium bisulfate irradiations completed
  - 5 LEU uranyl sulfate irradiations (2 – 30 hours)
- Phase-2
  - Most of the equipment has been fabricated
  - Experiments to begin in November 2015

Important System Components



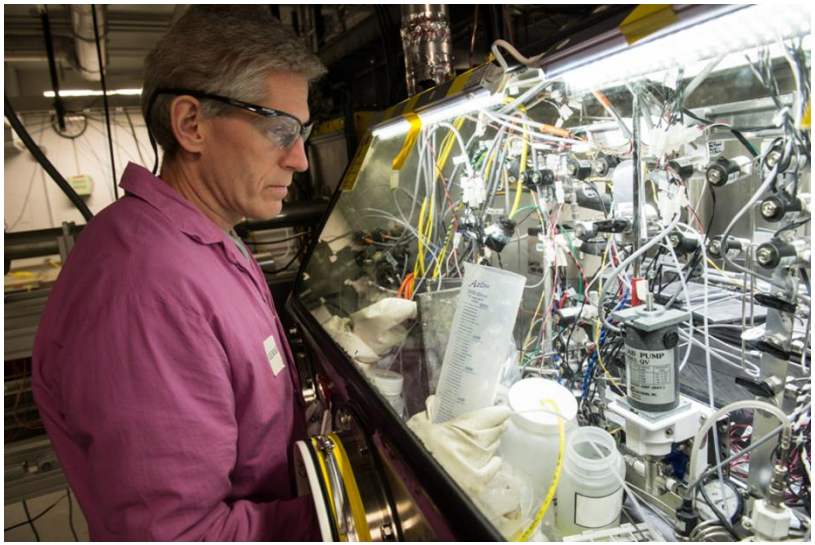
- 304 SS TSV with a 15-cm light-water reflector/cooler
- Shielded cell houses the TSV and Ta target
- Dump tank below shielded cell stores irradiated solution

Target Solution Monitoring Glovebox

- Up to 7 samples collected during irradiation – done remotely
- Bubbles prevented reliable use of pH, conductivity, and turbidity probes
- Samples retrieved 8-24 hours post-irradiation

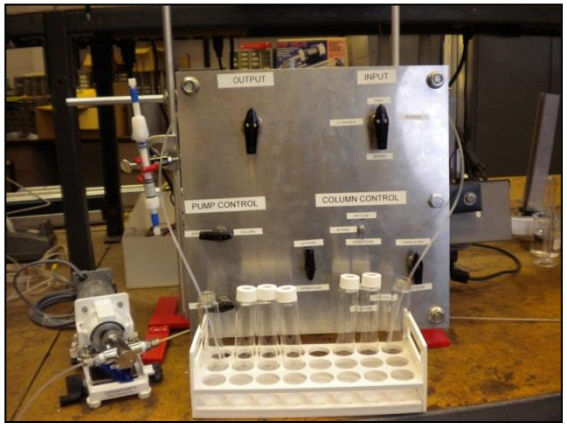
Mo-Recovery Glovebox

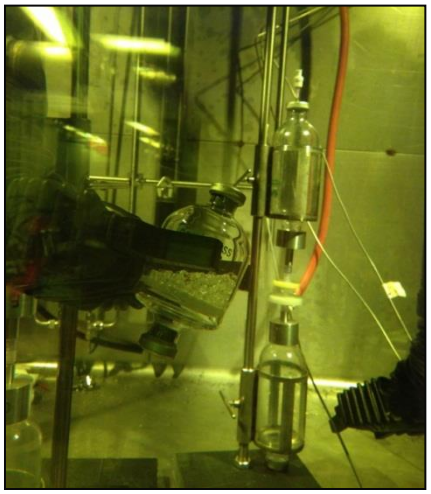
- Titania column to capture Mo-99 from irradiated uranium solution
- All operations are done remotely
  - Processing will begin 0-10 hours following irradiation
  - Target solution will be fed from the irradiation tank
  - Column effluent will go to the dump tank below the hot cell
  - Cold feeds are located inside the glovebox
  - Mo-product will exit the glovebox via a transfer line and go directly to 2nd hot cell for further processing
- Up to 15 samples can be collected from the feed, washes, and strip effluents
- Mo-product will be passed through a 2nd titania column and purified using the LEU-Modified Cintichem process



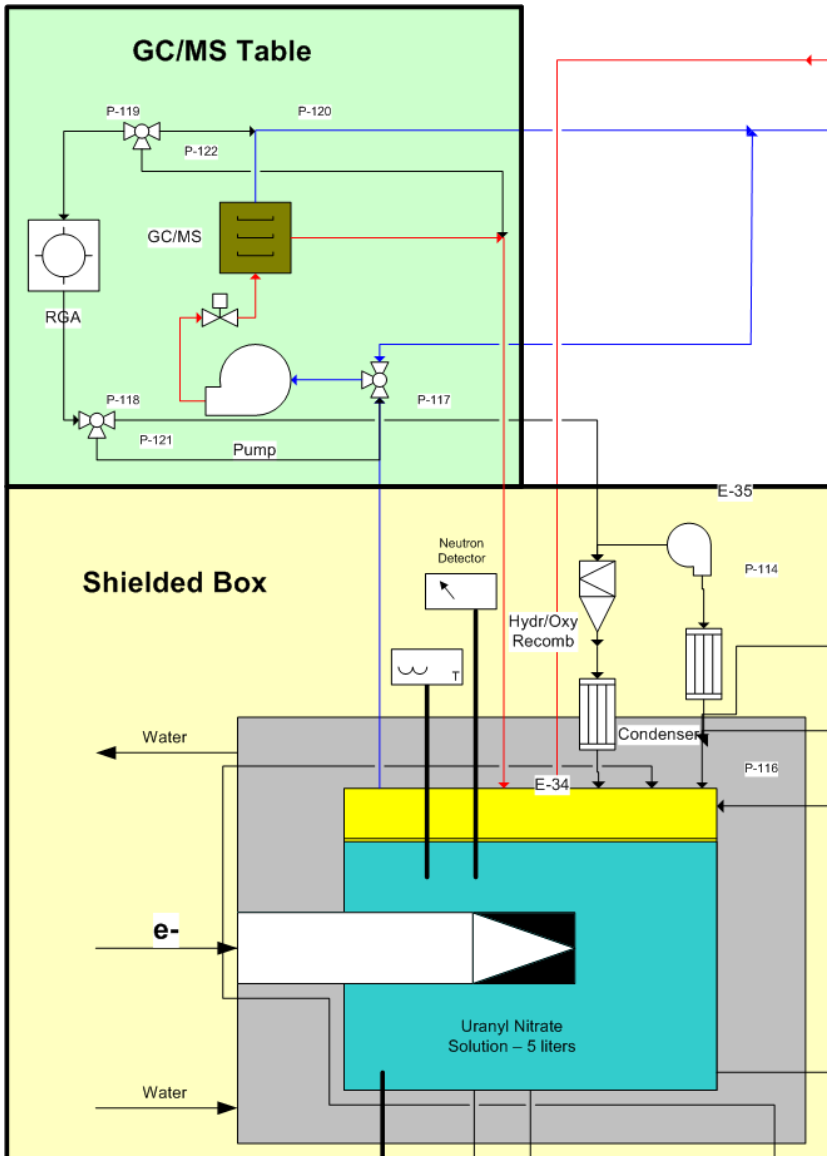
Concentration Column and LEU-Modified Cintichem

- In a second shielded cell (bigfoot), the Mo-product solution will be concentrated by a factor of ~15 using a much smaller column
  - Mo-product from the second column will then be acidified for entry into the LEU-Modified Cintichem process
  - Mo product will be concentrated down to 50 mL
  - LEU-Modified Cintichem process will be used to purify Mo-product



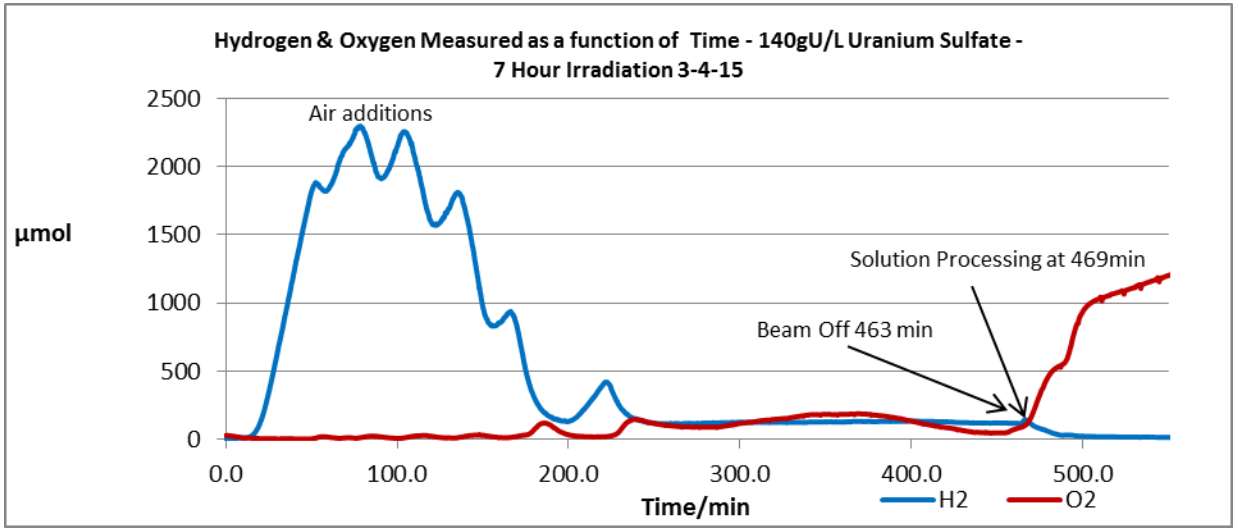


Gas Analysis System



Mini-SHINE Experiment—Off-Gas Analysis and Collection

- System is kept slightly negative by a 2-pump/3-tank off gas collection system
- Off gas in monitored by use of an RGA (Residual Gas Analyzer) during operation to measure hydrogen and oxygen generation
- A catalytic convertor is in-line to recombine hydrogen and oxygen
  - No oxygen generation has been observed for several hours after startup
  - Oxygen must be bled into the system during that time to keep H_2 level to below 1%
  - Samples of the off gas are collected and being sent to PNNL for analysis of volatile fission products
  - Thus far, it appears that the major fraction of radioiodine stays in the solution during operation



Gas Collection System

- All off-gases from experiment will be collected and decay stored
- Three cylinder system with increasing pressure <0=>4.5=>3500 psig
- Automatically maintain pressure in the solution vessel at -3 inches of water
- Final storage 6000 psig cylinder
- Pumps inside vessels to prevent pumps leaking into atmosphere

Mo-99 Purity Specifications

- **Goal is to produce 2 Ci Mo-99 that meets purity specifications for testing at GE Healthcare in the UK and 15- 20 Ci Mo-99 for testing at Lantheus Medical Imaging (phase 2)**

Ratio ($X/^{99}Mo$)	Product Specification
$^{131}I/^{99}Mo$	$\leq 5\times10^{-5}$
$^{103}Ru/^{99}Mo$	$\leq 5\times10^{-5}$
$^{132}Te/^{99}Mo$	$\leq 5\times10^{-5}$
^{89}Sr & $^{90}Sr/^{99}Mo$	$\leq 6\times10^{-7}$
$\Sigma\alpha/^{99}Mo$	$\leq 1\times10^{-9}$
$\Sigma\gamma/^{99}Mo$	$\leq 1\times10^{-4}$

- Total gamma results do not include Tc-99m, Mo-99, I-131, Ru-103, or Te-132 and have not been reported
- Total alpha results for Mo product met purity specifications all irradiations - ($<10^{-10}$ Ci-α/Ci-^{99}Mo)
- Ratios are based on activities 36 hours after EOB
- Te-132 was below detection limits for each final Mo-99 product
- Sr-89 & Sr-90 activities were based on Ba-140 activity which was below detection limits for each final Mo-99 product

Mini-SHINE Results

Irradiation	Time (hr)	Mo-99 produced (mCi)	Met Purity Specs[2]	Overall Mo-99 Yield
1	2	70[1]	Yes	95%
2	8	350	Yes	86%
3	32	810	No[3]	94%
4	20	380	Yes	42%[4]
5	12	190		

1. Insufficient mixing
2. Purity specifications do not include total gamma results
3. Purity specifications not met for Ru – change in base concentration on 1st recovery column – changed chemical form of Ru
4. Modifications made to Cintichem to help remove Ru from previous irradiation – longer contact with $KMnO_4$ – destroyed ABO-Mo complex

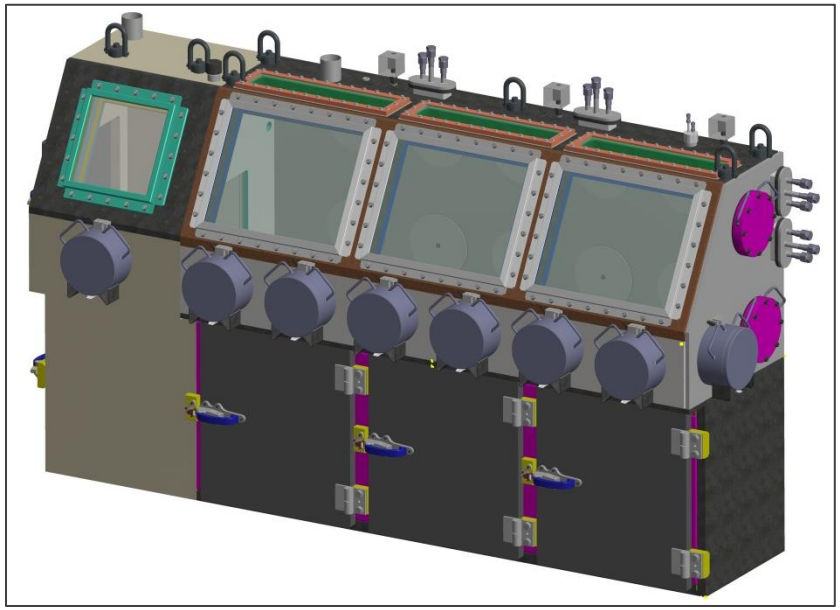
Irradiated Solution Chemistry

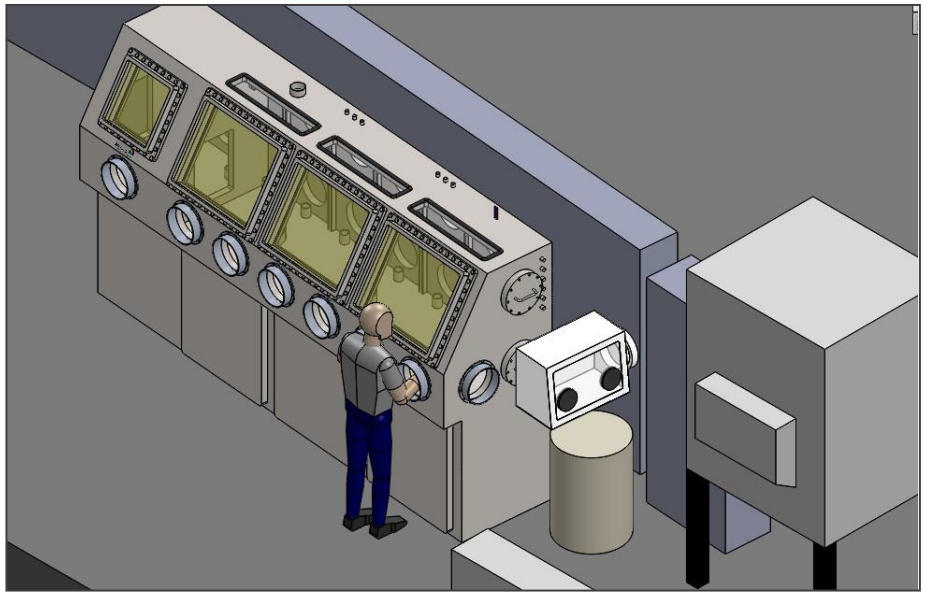
- No changes in redox chemistry for Mo-99 (phase 1 conditions)
- Only ~30% Te remained adsorbed on 1st titania column
- >90% Zr remained adsorbed on 1st titania column
- 1st titania column – Ru(40%), Ce(15%), and Sb(5%)
- Fission products that co-eluted with Mo-product from 1st titania column – Ru, I, and Sb
- No precipitation of fission products or formation of uranyl peroxide

What's Next?

- Experiments have been delayed due to decrease in U concentration – dilution event in dump tank & spill
- Experiments to resume next week
- LEU solution will be reconstituted(94 g-U/L → 140 g-U/L)
- 2-hour irradiation will be performed to get new production rates
- 2 Ci production run with shipment to GE Healthcare in mid-September
- Few short irradiations will be performed – micro-SHINE – peroxide formation experiments
- Phase 1 will be removed for phase 2 installation

Phase 2 Mini-SHINE Experiments





- Phase 2 will have a single glovebox
- Windows are Pb-glass to provide additional shielding
- Cold solutions will be kept below the interior of the box
- Transfer port will be used to bring materials and samples in and out
- Valving system will be similar to phase 1 – solenoid valves
- Column will be larger (3.5 cm ID X 13 cm L)
- Flow rates will be ~170 mL/min
- CV is 125 mL

Summary and Conclusions

- Mini-SHINE experiments - mini-pilot plant for SHINE
- Chemistry is good – no changes in redox at least for Mo-99
- After ~3-3.5 hours of irradiation, hydrogen and oxygen reach a steady state
- Purity specifications met for I-131, Ru-103, Te-132, and Sr-89/90 for all irradiations except 3rd
- Important shipment to GE Healthcare in UK in September 2015
- Important shipment to Lantheus Medical Imaging (December 2015)

Acknowledgements

- The submitted manuscript has been created by UChicago Argonne, LLC, Operator of Argonne National Laboratory (“Argonne”). Argonne, a U.S. Department of Energy Office of Science laboratory, is operated under Contract No. DE-AC02-06CH11357. The U.S. Government retains for itself, and others acting on its behalf, a paid-up nonexclusive, irrevocable worldwide license in said article to reproduce, prepare derivative works, distribute copies to the public, and perform publicly and display publicly, by or on behalf of the Government.
- Work supported by the U.S. Department of Energy, National Nuclear Security Administration's (NNSA's) Office of Defense Nuclear Nonproliferation, under Contract DE-AC02-06CH11357.