Mo-99 Production from a Low Enriched Uranium Sulfate Solution:

The Blue Room Experiment

A.S. Anderson, L.J. Bitteker Jr., M.A. Connors, R. Copping, M. Cover, W.J. Crooks, G.E. Dale, D.A. Dalmas, M.J. Gallegos, E. Garcia, J.G. Gioia, R. Gonzales, D. Graves, W.K. Hollis, M.T. Janicke, C.T. Kelsey IV, I. May, M. Mocko, M. Pieck, M. Rawool-Sullivan, S.D. Reilly, D. Rios, T.J. Romero, F.H. Stephens, F.L. Taw, D.L. Thorn, and K.A. Woloshun

Los Alamos National Laboratory
Los Alamos, NM 87545, USA

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“Blue Room” Irradiation and Chemistry – Project Goals

- **Aimed for ~ 100 mCi production of Mo-99 (2 orders of magnitude greater than previous irradiations) from LEU solution**
- **Design, build, test, and use gas handling apparatus during irradiation**
  - Catalytic converter for recombination of H_2 and O_2 generated from radiolysis
  - Iodine trap
  - Residual gas analyzer mass spectrometer for real-time analysis of gases
- **Demonstrate separation chemistry in a hot cell**
  - Design, build, test, and use equipment to be compatible with hot cell manipulators
  - Hot cells located in a Category II Nuclear Facility (CMR)

Previous Irradiations Performed

- **March 2011 Linatron Experiment (NISC)**
  - Two 70 mL samples, light and heavy water uranium nitrate
  - Produced ~ 20 μCi ^{99}Mo total
- **May 2011 Linatron Experiment (NISC)**
  - Three 13 mL samples, LEU nitrate and sulfate samples
  - Produced ~ 3 μCi ^{99}Mo each
- **June 2011 Blue Room Experiment**
  - Four 8 mL natural U nitrate and sulfate samples
  - Produced ~ 300 μCi total
- **December 2011 Linatron Experiment (NISC)**
  - Re-irradiated the three 13 mL samples from May
  - Produced ~ 3 μCi ^{99}Mo total
- **January 2012 Blue Room Experiment**
  - Four ~12 mL LEU nitrate and sulfate samples
  - Produced ~ 1.4 mCi ^{99}Mo total
- **December 2012 Target 4 Experiments**
  - Iterative irradiations of LEU solutions
  - Produce ~ 1 mCi ^{99}Mo

February 2013 Blue Room Experiment Parameters

- **300 mL of 150 g(U)/L uranium sulfate (19.54% enriched), pH 1**
- **Irradiation parameters:**
  - 88.3 hours of irradiation
  - 2.17E+17 protons were delivered to W target
  - Average beam current = 109 nA
  - Highest beam current = 139 nA (instantaneous reading)
  - Total thermal neutron fluence in the reaction vessel = 3E+15 n/cm^2
- **Radiolytic gas production - 0.88 mL/min for H_2 and 0.44 mL/min for O_2**
- **Reactor operating temperature ~ 26 ^{o}C**
- **Operating pressure < 15 psia**
- **Oxygen < 3% by volume (diluted with argon)**

Major Safety Considerations

- Radiation (dose to workers, degradation of materials, electronics failures)
- Radioactive contamination from breach of systems (workers, equipment, facility, environmental release)
- Corrosive and toxic samples (LEU, pH 1 acidic solution)
- Flammable gas mixtures (hydrogen and oxygen produced by radiolysis)
- Pressure build-up
- Temperature excursions
- Equipment failure
- Security Consideration: Accountable quantities of ^{235}U (> 0.5 g)

Conceptual System Design P&ID

MAWP=7 PSIg Circulation
MOP=15 PSIA recombiner/condenser

Drawing of February 20, 2013

Comments:

1 Heat Tape Controller: Watlow EZ-Zone PM Controller
2 Mass Flow Meter: Sage Metering Inc. SRP-025-TUBE-VCR025-RA25-DC24-AR-H2
3 Mass Flow Controller: Horiba Z500 MFC 4-20mA
4 Air Solenoid Valves: SMC Series VZ215-5LZ-01 (Not shown)
5 Diaphragm Pump Controller: DATEXEL 5022; 0 - 20mA to 0 -5V Converter
6 Diaphragm Pump: TBD
7 Catalytic Converter: Resource Systems Inc. Model RS-4889A
8 Heat Tape OmegaLux FRG-030 120VAC 125W 3ft length
9 Quick disconnect: Eaton MLDB series (female: ML2DBS25F292, male: ML2DBP25F292)
10 Condenser
11 Pressure transmitter: IMP-A-2000-1A4-CSV-XXX-000
12. Thermocouples: Type J
13. Iodine Trap: In-house design (Ionex Ag exchanged zeolite X)
14. Vacuum Sensor: Baratron or Equiv.
15. Swagelok Ball Valve SS-63PSW8T-33C-L12839

Note: All AVs will use ¼" air line from air solenoid to bellows valve.

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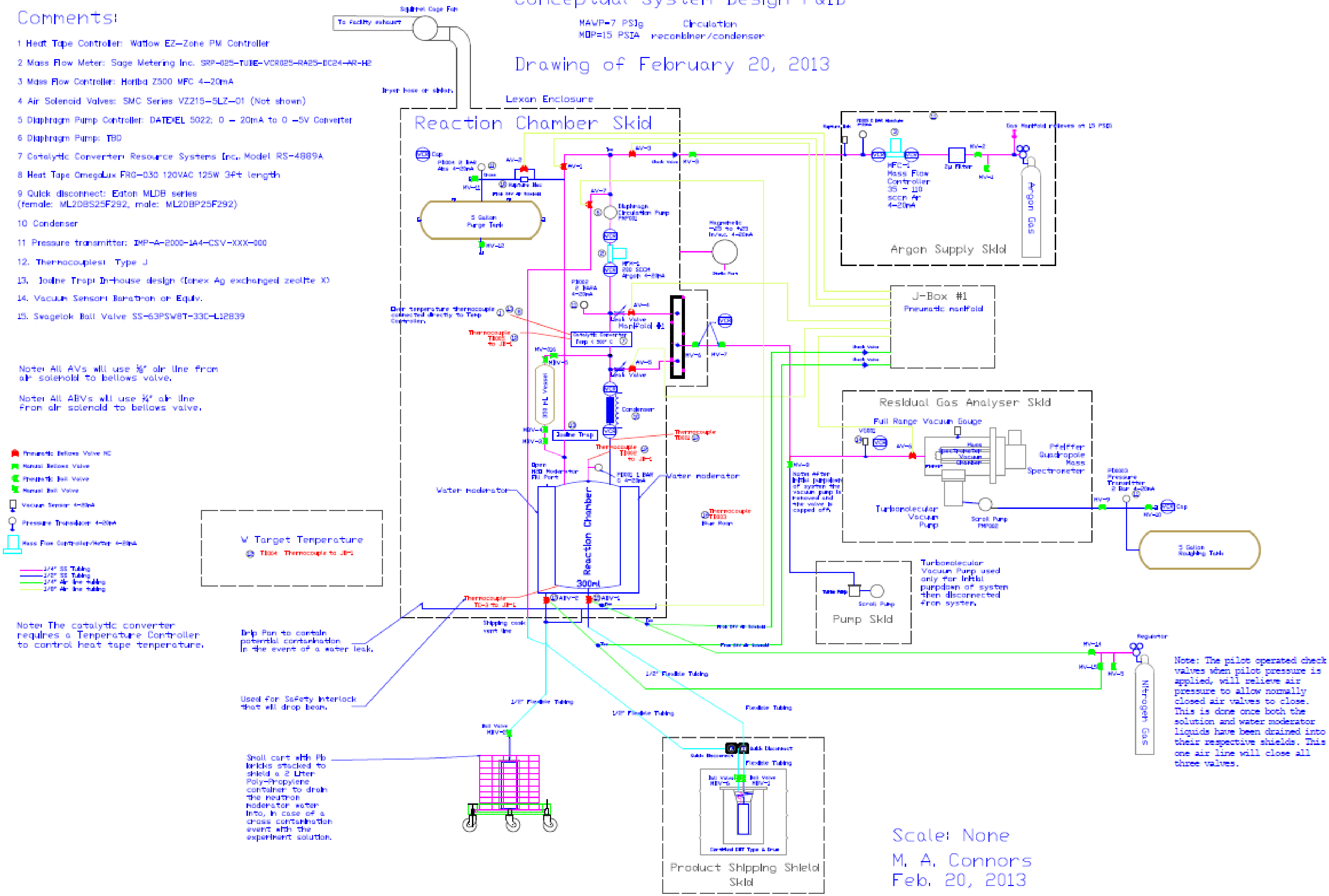
Note: The catalytic converter requires a Temperature Controller to control heat tape temperature.



Scale: None

M. A. Connors
Feb. 20, 2013

Gas Handling System

Moving the Gas Handling System to the Blue Room

Installed in the Blue Room

Control System During Irradiation

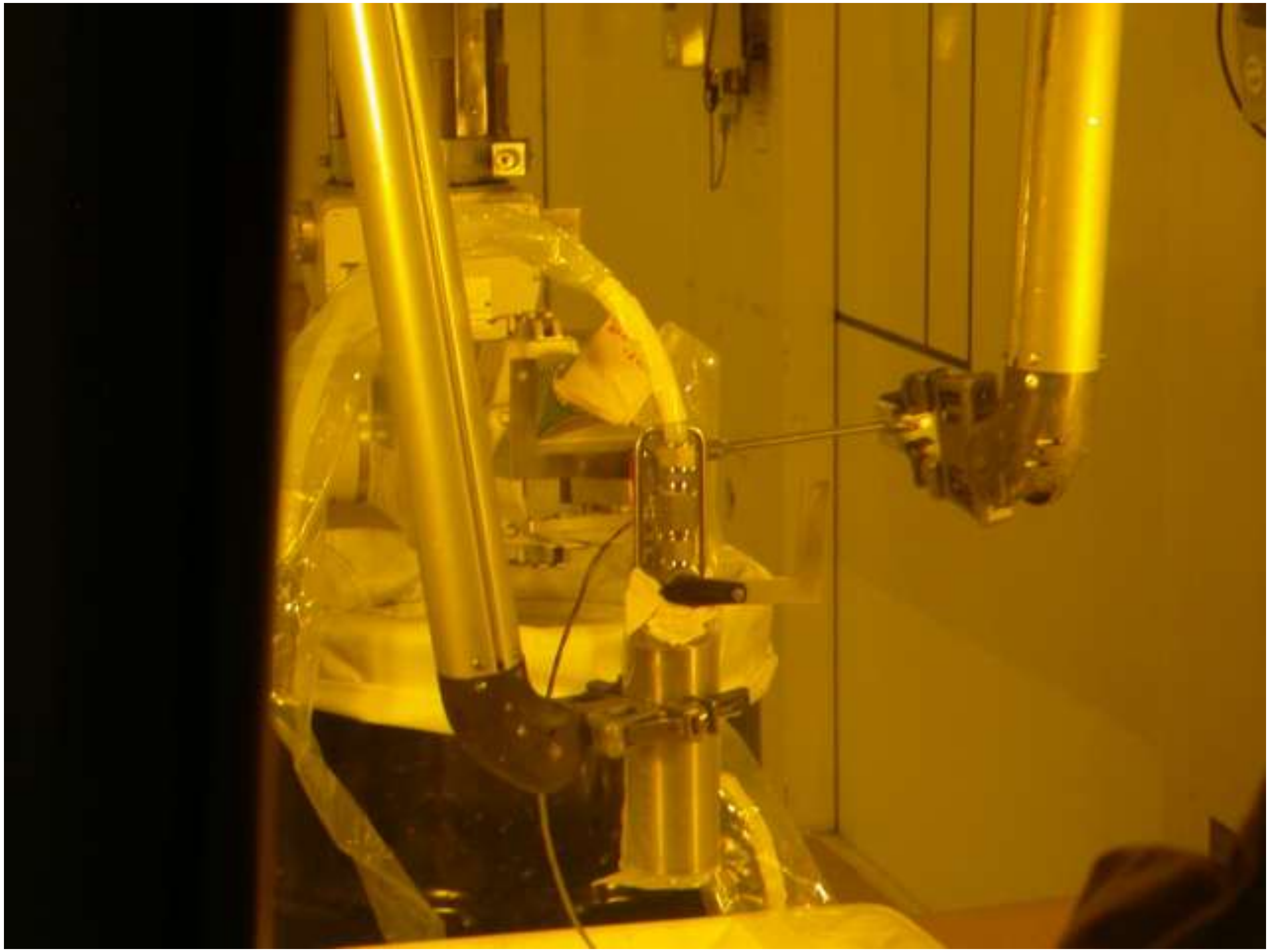
RGA Mass Spectrometer Used for Real-time Analysis

Draining Irradiated LEU to Transport Cask

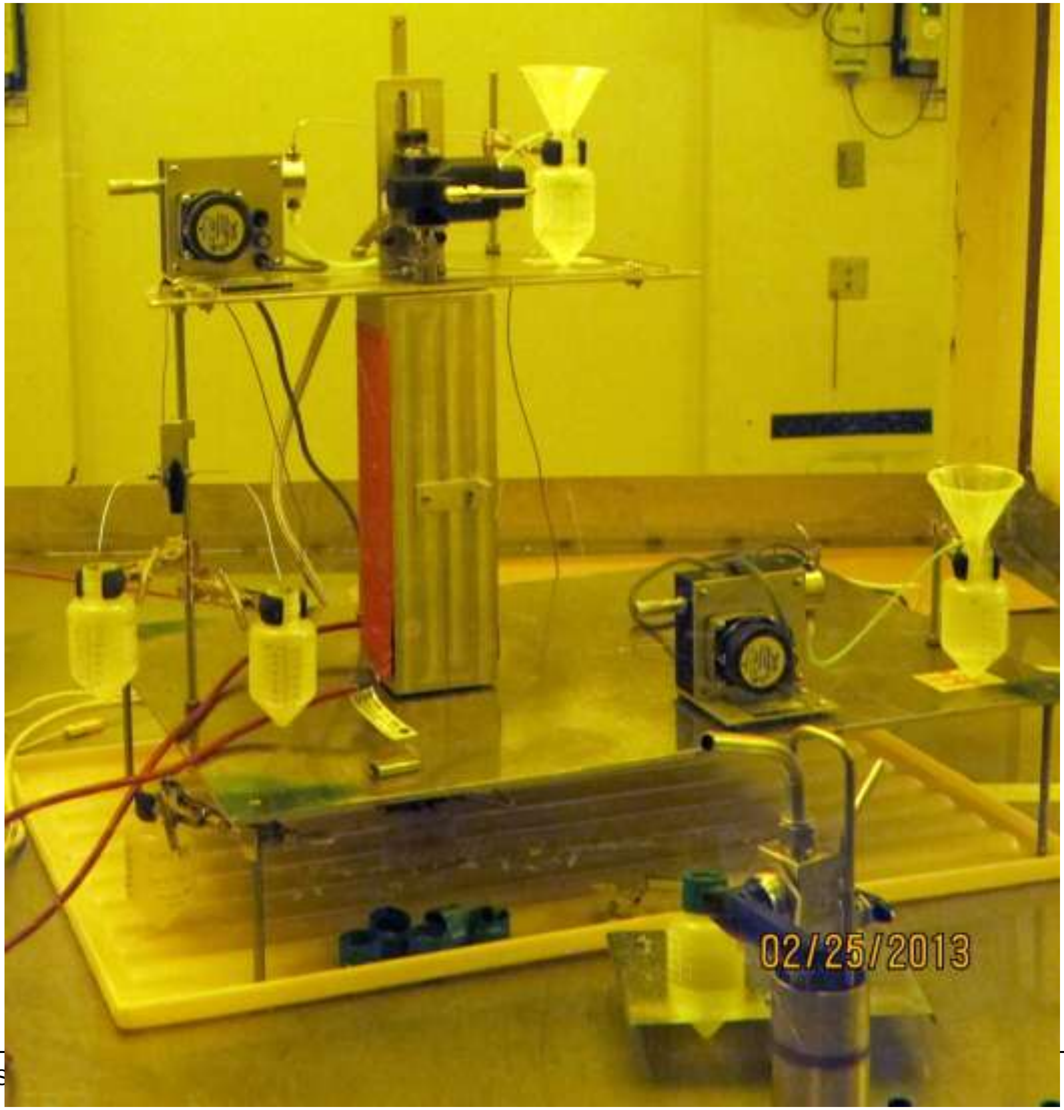
Lifting the inner drum out of the 9979 shipping drum

Opening the inner shipping drum inside the hot cell corridor

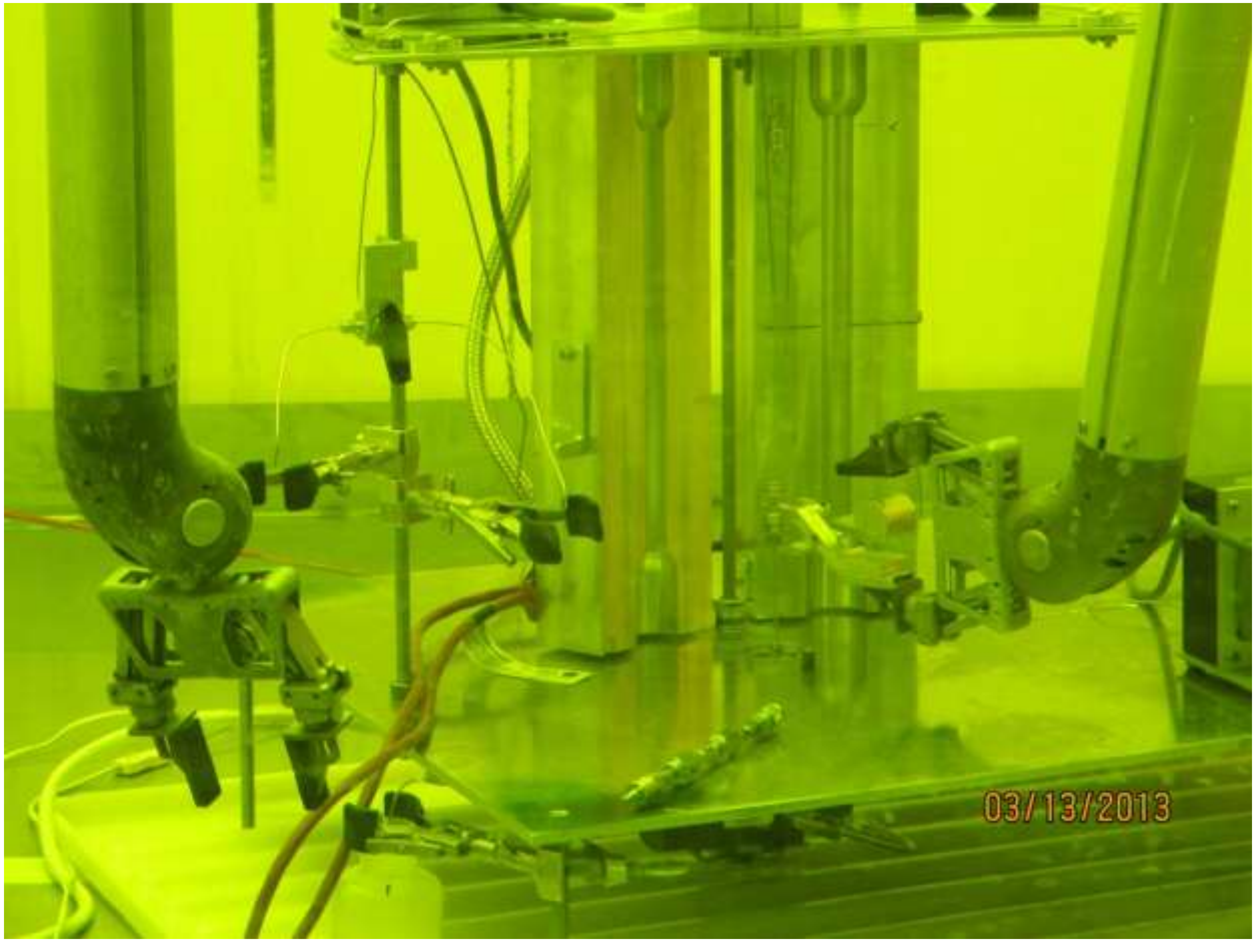
Using manipulator hands and a wrench to detach the drain tube from the shipping vessel in the hot cell corridor



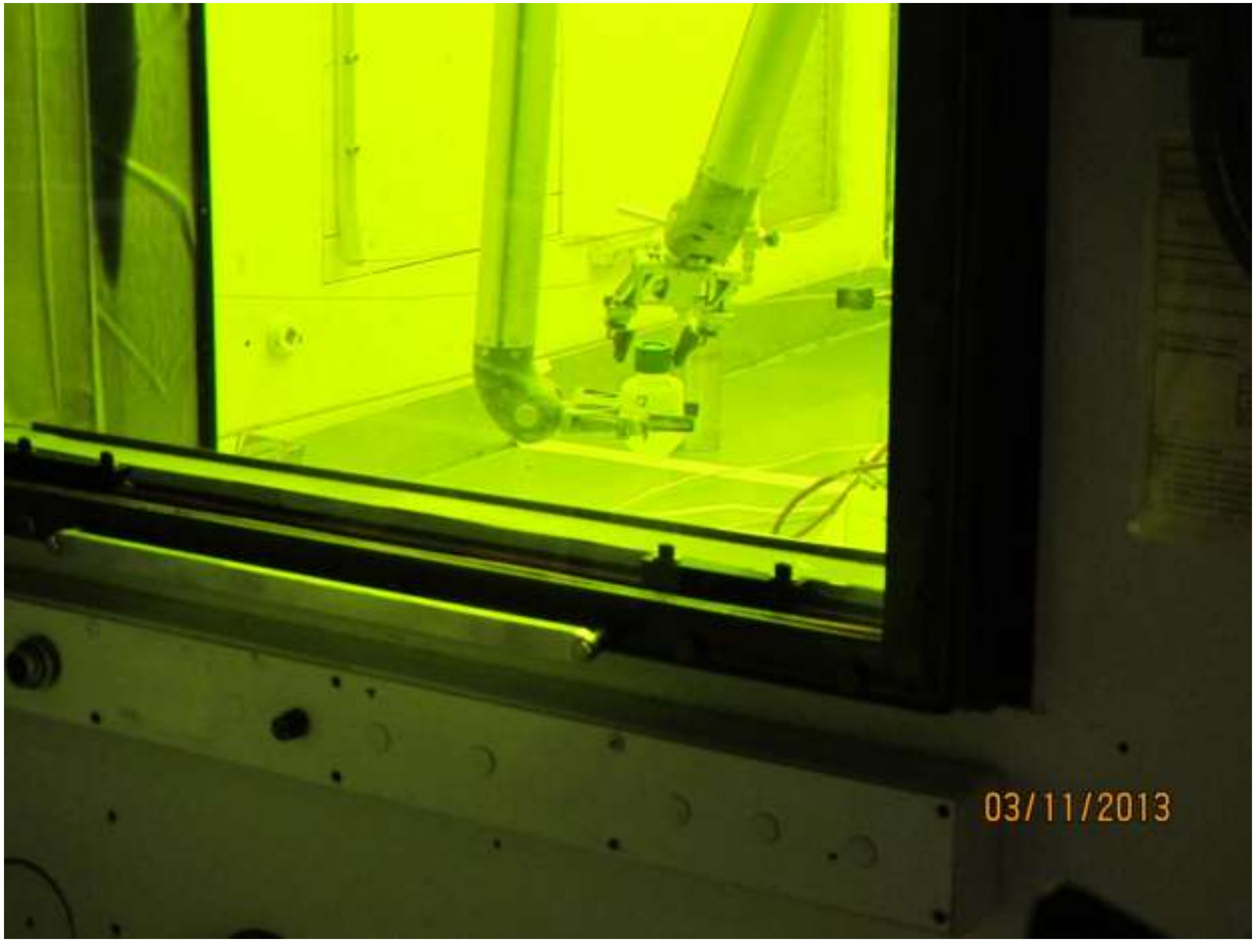
Separation apparatus



A view of the heating block (open) and the specially made tool for actuating miniature quick connects with the manipulator hand (right). The column with its quick connects is laying on the stand's lower shelf (bottom center).



Using the manipulators to uncap a bottle



Fuel Parameters Pre- and Post-Irradiation

- **~ 6 mL of liquid remained in gas handling/reaction vessel system in Blue Room after draining to transport vessel (indications point to pure water loss, not LEU)**

Fuel analysis	Before Irradiation	After Irradiation
Sample mass	358.46 g	352 g
Density	1.192 g/mL	1.195 g/mL
Uranium concentration	150.0 gU/L	155.3 gU/L
Solution pH	0.93	1.03

Mo-99 Production at End-of-Beam

- **Preliminary results from 3 independent methods show ~ 70 mCi production of Mo-99**

Isotope	Production values at TA-48	Production values at CMR		Production value based on Gold foils
Mo-99 (181 keV)	67(8) mCi	70.00(1.07) mCi		67 mCi
Mo-99 (740 keV)	57(1) mCi	69.05(0.97) mCi		

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Mo-99 Separations

- **Irradiated solution split into two ~145 mL aliquots and one ~10 mL aliquot**
- **145 mL aliquots were used for separations through titania**
- **Next presentation (Iain May, LANL) will provide details of separations**

% recovery of Mo-99	181 keV	740 keV
First titania column	85%	95%
Second titania column	81%	90%

Accomplishments from the Blue Room Experiment

- **Produced ~ 70 mCi production of Mo-99**
- **Designed, built, tested, and used gas handling apparatus during irradiation (many lessons learned!)**
  - Catalytic converter for recombination of H_2 and O_2 generated from radiolysis
  - Iodine trap
  - Residual gas analyzer mass spectrometer for real-time analysis of gases
- **Demonstrated separation chemistry in a hot cell**
  - Designed, built, tested, and used equipment to be compatible with hot cell manipulators
  - Hot cells located in a Category II Nuclear Facility
- **Next experiment – produce 8 Ci of Mo-99 (approximately 100x scale-up)**