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2012-13 Blue Room Low Enriched Uranium Sample Irradiation, Associated Gas Handling System, and Subsequent Separation Chemistry

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Executive Summary

An irradiation experiment was designed for the production of 100 mCi of medical isotope Mo-99 from a Low Enriched Uranium (LEU) solution. 300 mL of 150 gU/L LEU sulfate solution (pH1) was prepared and irradiated at the LANSCE Blue Room facility. The apparatus designed for the irradiation included a gas handling system, in which safe operation could be controlled by monitoring change in gas pressure. The gas handling system also contained a hydrogen oxygen recombiner, to recombine the gases generated through radiolysis of water, and an iodine trap, primarily for I-131. Pressure transients were observed during irradiation, which could be controlled by increasing the flow rate of the circulating pump. After irradiation the LEU solution was transferred to a hot cell facility for subsequent Mo-99 recovery using a titania column separation process, with samples also subsequently shipped to a radiochemical laboratory for further analysis. Gamma spectroscopy analysis revealed that ca. 70 mCi of Mo-99 was present at end of beam, in good agreement with the predicted value when actual beam history was included into the calculation. There was 80-85 % recovery of Mo-99 through a titania column separation, excluding losses through radioactive decay. The % Mo-99 recovery was lower than observed previously for the 1 mCi Mo-99 production (Target 4) experiments undertaken at LANL using the same separation apparatus, with on this occasion evidence of lower oxidation state molybdenum remaining irreversibly bound to the titania column. Subsequent post mortem analysis of the Blue room apparatus revealed that there had been no leaks in the gas handling system, the hydrogen and oxygen recombination appeared to work efficiently, and recovery of irradiated uranium trapped in the system allowed for >99% uranium mass balance.

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1. Introduction

Previously, in support of SHINE Medical Technologies^{TM}, we have irradiated multiple small scale samples and provided technical support for the first stage of the ^{99}Mo recovery process from irradiated Low Enriched Uranium (LEU) target solutions. This work was funded through the NA21 Global Threat Reduction initiative (GTRI). Separation chemistry experiments indicated that ^{99}Mo could be recovered from the vast excess of LEU and many of the fission products present in solution using titania as a selective ^{99}Mo sorbent. In FY13, to test the titania column separation process on a direct downscale of process operation, 150 mL of LEU sulfate solutions (pH 1) were irradiated to produce 1 mCi ^{99}Mo using new sample containment methods and a new irradiation capability at Target 4 (LA-UR-13-28967). The bulk of the irradiated solution was then passed through titania column separation apparatus designed at LANL using operating conditions designed by Argonne National Laboratory (ANL) - based on 'VERSE' code simulation developed at Purdue University. Near quantitative recovery of ^{99}Mo was obtained, even when uranium recycled from a previous separation experiment was re-irradiated and the ^{99}Mo titania separation performed on this recycled fuel.

In the course of the whole suite of LEU irradiation experiments undertaken in FY11-13 gas containment, handling and analysis methods were improved and refined. However, to more accurately reflect projected SHINE process operation, higher production densities of ^{99}Mo must be produced. During the production of higher activities of ^{99}Mo the build-up of radiolysis gases (H_2 and O_2) from radiolysis of water necessitates the use of an on-line gas recombination system. In addition, moving beyond 1 mCi of ^{99}Mo increases radiation doses to a level that makes it impossible to undertake separation chemistry in a fume hood, where all separation chemistry analysis has been undertaken thus far. Thus separation chemistry for irradiations > 1 mCi must be performed in a hot cell.

By May '12 a LANL team was being assembled to undertake an experiment (called the "Blue Room Experiment") to produce up to 100 mCi of ^{99}Mo from an LEU sulfate solution over a 5 day irradiation, with subsequent recovery of ^{99}Mo from the irradiated solution using the titania separation process. This required the design and construction of a gas handling system that could both recombine H_2 and O_2 and trap radioactive iodine isotopes (primarily ^{131}I), as well as the design of titania column separation apparatus applicable to hot cell operation. This endeavor necessitated the coordination of efforts between three facilities; the radiochemistry laboratories at Technical Area (TA) 48, the neutron irradiation capability at the Blue Room, LANSCE (and associated LANSCE cold space) and the Chemistry and Metallurgy Laboratory (CMR) hot cell facility. Subsequent additional NA-21 support allowed for decommissioning of the Blue Room irradiation apparatus back at TA48, and subsequent post-mortem analysis of key components. A schematic outlining the facilities where different operations were conducted, and the timeline for these different operations, is presented in Figure 1.

2. Target Solution Preparation and Shipments

The LEU sulfate target fuel used for this Blue room experiment was exactly the same LEU sulfate fuel as used for the Dec. '12 – Jan. '13 Target 4 experiments (soln. density - 1.192 g mL^{-1}, pH 1.0, 150(1) gU L^{-1}, 19.54 % ^{235}U). Prior to loading the 300.7 mL of target solution into the transportation cylinder, it was spiked with natural molybdenum (106 µL of a 10.08 mmol L^{-1} $Na_2MoO_4.2H_2O$ aqueous solution) as to mimic the molybdenum concentration at the proposed SHINE production levels (3.55×10^{-5} mmol L^{-1} of natural molybdenum). Iron was also added to the fuel (879 µL of a 0.031 mol L^{-1} $Fe^{III}_2(SO_4)_3$ soln.) to decompose any hydrogen peroxide that could have been produced during fuel irradiation. Conversations with M. Youker (ANL) indicated that both Fe(III) and Fe(II) would catalytically decompose any radiolysis generated peroxide, with the higher oxidation state of iron chosen to minimize the risk of reducing Mo^{VI} to lower oxidations states of Mo (which would impact the subsequent separation chemistry). After the two chemical additions, the resultant doped LEU solution was swirled and then transferred into a 500 mL squirt bottle which had been modified with a long dispensing tip. The LEU sulfate target fuel was then dispensed into the stainless steel cylinder used for fuel shipment to the LANSCE Blue Room facility, with the long tip on the squirt bottle allowing for the solution to be added directly into the cylinder without contacting the valve area. The stainless steel cylinder was then transferred to the gas handling fume hood and connected to the gas manifold where the head space in the cylinder was purged with argon for 1 min. After purging with Ar the cylinder was then disconnected from the manifold and endcaps were attached to both valve openings on the cylinder. The cylinder was then brought out of the hood and the Swagelok handles of each valve removed to prevent them from accidentally being opened during shipment. The radioisotope inventory contained in the cylinder, from the LEU sample, was written on yellow tape and stuck on the outside of the cylinder. The cylinder was then packed inside a 30 gal drum which was subsequently contained inside a special DOT certified 55 gal. drum (type 9979) and shipped to LANSCE.

Prior to irradiation, the transport cylinder containing the target solution was attached to the reaction vessel and the lower valve opened, draining the solution into the reaction vessel. After irradiation, the now irradiated LEU solution was drained from the reaction vessel into a new transport vessel that had been pre-loaded into the inner shipping drum, for transfer to the CMR hot cells. The drum contained lead shielding to meet packaging and transportation requirements. The inner shipping drum was loaded into a special DOT certified 55 gal. drum (type 9979) and sealed. The dose rate measured at contact of the inner drum lid was 35 mR/hour.

3. Radiolytic and Radioactive Gas Handling

This section describes the experimental portion of the gas handling system built and used for the production of ^{99}Mo from the LEU target solution in the Blue Room at LANSCE. Many considerations, discussions, and design modifications over the course of 8 months resulted in the final system. Reoccurring concepts with regard to safety and successful operation included material compatibility with the radiation fields, minimization of gas volumes, maintaining hydrogen concentrations below flammable limits, operating at close to atmospheric pressures, and the ability to fill and purge the system remotely without releasing gases to the ventilation stacks. This section will describe the gas handling system, pressure calculations used to work in a low pressure safety envelope, operation during the LEU irradiation, results and lessons learned.

3.1. Description of the Blue Room Apparatus, Focusing on the Gas Handling System

The system can be briefly described as a closed loop gas flow with sampling feeds to a remotely located residual gas analyzer (RGA)/mass spectrometer, a fill line from an argon supply, and a large ballast tank into which the final gas could be purged. The precise details were incorporated into a Piping and Instrumentation Diagram (P&ID). The closed loop nature of the system prevented radiolytic gases from being vented into the emission stacks at LANSCE. Appropriate plumbing (VCR and Swagelok fittings) and rupture discs preserved the low pressure operation of the system. Contained within the loop were units to maintain flow, monitor pressure, remove radioactive iodine, recombine radiolytically produced H_2 and O_2 in a heated catalytic converter, and a small heat exchanger to return the gas to the reaction vessel. Thermocouples were also added to monitor the temperatures for the reaction vessel, recombiner, gas temperature leaving the condenser, and the Blue Room ambient temperature. A gas flow rate of *ca.* 70 sccm maintained a H_2 vol. % of less than 1% based on calculations of 0.66 sccm H_2 produced from radiolysis. Shown in Figure 2 is the flammability diagram for mixtures of hydrogen, oxygen and nitrogen. From this data it is seen that for mixtures below 5% O_2, the gas does not enter into the flammable regime. Subject matter experts slightly shift this value for argon (vs. nitrogen) to closer to 3% O_2. With concentrations of 1% H_2 and a maximum of 0.5% O_2 by stoichiometry, the system was well below the flammable limit for the predicted gases produced.

The Blue room apparatus was built on to a trolley, with other major components including a stainless steel reaction chamber, stainless steel solution transfer vessels, a stainless steel purge tank, Be reflectors, and lead shielding. The apparatus was built on to a 80/20 modular alumina frame and enclosed by panels of Lexan plastic. Schematic diagrams and a picture of the apparatus are shown in Figure 3 and Picture 1, respectively.

3.1.1. The Iodine Trap

The main concern for the iodine trap was to prevent I_2 from reaching the RGA outside of the Blue Room, and to protect the Pt/Pd catalyst in the O_2 & H_2 recombiner. It is known that I_2 can react with the metal catalyst and create volatile species that can degrade or poison the recombiner. For the trap, 2.5 g of Ionex silver exchanged zeolite (Picture 2) was placed in a ½" stainless steel tube and kept in place with glass wool plugs. This assembly with VCR fittings was then installed into the system. Ag exchanged zeolites have a capacity of roughly 100 mg I_2/gram zeolite, thus there is a clear excess of zeolite and capacity should not have been reached ("Methods of Gas Phase Capture of Iodine from Fuel Reprocessing Off-Gas: a Literature Survey", Haefner and Tranter, February 2007). The zeolite was also fully conditioned in the atmosphere to absorb water such that an exothermic absorption event would not occur, as had been reported for a nuclear reactor in 1986 (U.S. NRC Information Notice No. NO 86-43).

3.1.2. Diaphragm Recirculation Pump

A small Dia-Vac diaphragm pump (Air Dimensions Inc., single head B-series), with 316SS wetted parts and Viton polymers, was installed to recirculate the gas flow. In general, fluorinated polymers were excluded from the system; however, the distance from the reactor, the nature of the Viton, and exposure calculations indicated that it was an acceptable choice for this pump. This was an exceptionally quiet pump and the speed was controlled remotely with a variable power supply (Picture 3).

3.1.3. Mass Flow Meter and Pressure Transmitters

A Sage in-line Thermal Mass Flow meter and Impress industrial pressure transmitters with piezo-resistive ceramic pressure sensors were used for remote mass flow monitoring and pressure recording, respectively. Both systems were selected to be appropriate for the radiation fields and performed well. A lesson-learned, however, was discovered with the mass flow meter. Due to the pulsating nature of the diaphragm pump, the meter could not accurately measure the flow and would report a fixed higher flow at the meter's maximum value during circulation. The meter would also report a low flow without circulation. This was later determined to be due to the meter's dependency on the gas composition, especially the amount of hydrogen in the argon cover gas, which changed during the experiment. This was not deemed a safety critical component for operation so it was used as a simple flow detector instead of a measuring unit.

3.1.4. Recombiner for H_2 and O_2

A recombiner was used for the catalytic conversion of H_2 and O_2 produced during radiolysis of water, thus maintaining the uranium concentration and pH in the LEU solution in the reactor by returning the H_2O_{liq} to the system. The recombiner was purchased from Resource Systems, Inc (Model RS-4889A) and had 5.3 in^3 volume with 400 cells/in^2 (Picture 4). The honeycomb support was impregnated with 0.5 wt% metal in a 2:1 stoichiometric ratio of Pt:Pd. It was heated to 200 ^{o}C with external heating tape to maintain a temperature above the boiling point of water to avoid condensation issues. A thermocouple attached to the recombiner was monitored to ensure the proper operating temperature. Unexpected heat losses from the reactor were observed and additional heating from the H_2 and O_2 recombination (-57.9 kcal/mol) was not observed. A miniature heat exchanger from Exergy (10 series with 7 tubes in shell and tube design, ca. 7 inches in length) was installed to further cool the gas before reintroducing the flow to the reaction chamber. At this point the heat exchange was minimal; however, future designs with higher flow and heat production rates could use the heat exchanger for cooling.

3.1.5. Gas Sampling and Residual Gas Analyzer

After the iodine trap, and before and after the catalytic recombiner, Granville-Phillips Variable Leak Valves (from Brooks) were installed to send small samples through an evacuated piping system to a Stanford Research System (SRS) RGA-100 mass spectrometer (Picture 5). The system involved over 50 feet of 3/8" stainless steel piping connected with small bellows sections and Conflat flanges with copper gaskets. A vacuum of 10^{-6} torr was achievable with a scroll pump for rough pumping and a large turbopump. The decision was made to sample the gases after the iodine trap to prevent long-lived iodine species leaving the experimental apparatus in the Blue Room, and in any case calculations show all volatile fission product isotopes would inevitably be below the detection limit of the RGA. An RGA with a mass limit of 1 to 100 amu was selected for higher precision monitoring of the gas concentrations of Ar, H_2, O_2, N_2 (if a leak was detected) and H_2O. Due to the unknown lag time, sensitivity and longevity with the RGA, the concentration of H_2 in the gas phase measured with this system was not used as a part of the safe operation basis.

3.2 Safety Considerations during Irradiation

As mentioned above, the concerns for the safe operation of the gas handling system included staying below both the hydrogen flammability limit and the maximum operating pressure for the reactor. The team was unable to find a reliable H_2 sensor appropriate for the high radiation field, so it was decided that three critical monitors were needed for operation: at the heater for the recombiner to monitor operating temperature (section 3.1.4.), flow indication from a mass flow meter (section 3.1.3.), and most critically pressure monitoring of the gas system (section 3.3.3.). Calculations show that if the recombiner or pump failed and the pressure increased from gases produced from the radiolysis of water, it would be indicated by an increase in the overall system pressure. Shown in Figure 4 are the calculations for the pressure increase with time and the associated H_2 and O_2 concentrations ***if*** the catalytic recombiner failed to convert hydrogen and oxygen to water. With this model, the system starts at 2 psig and hydrogen and oxygen initially remain dissolved in the aqueous LEU solution. As time progresses, H_2 and O_2 evolve as ideal gases and the pressure increases. What is observed in this calculation is that there would be roughly a 20 min window of time with which corrective action could be taken if a pressure increase was observed. Such corrective action would include turning the beam off, the main driver for radiolysis reactions, to stop any pressure excursion. The system was also designed to allow for the addition of argon to compensate for the slow leak to the RGA for analysis.

3.3. Results

The LEU solution was successfully irradiated for over 6 days with several interruptions arising from off-normal events. These events were unexplained pressure increases, and a brief explanation of possible causes will be discussed later. The results presented here are limited due to a limited ability to monitor gas concentrations with the RGA, and observed temperature changes that did not reflect the chemical reactions in the system. Nevertheless, this was a very well planned experiment where slight changes in the monitored pressures acted as a direct tool to ensure operation within the safety envelope.

3.3.1. Residual Gas Analyzer Mass Spectroscopy Results

As previously stated, the team was unable to incorporate either a H_2 or an O_2 sensor into the design in the relatively short amount of time available to the preparation of the gas handling system. This was mainly due to presence of self-contained electronics that would not be stable in the radiation fields. For this reason the RGA was used to monitor the gas concentration; however, it did not perform as a real time measurement as hoped. The instrument was calibrated with a 1% H_2 and 0.5% O_2 by volume. However, during the course of the experiment, O_2 was below the detection limit of the RGA and the sensitivity towards H_2 seemed to increase. Changing from the Faraday detector to the Electron Multiplier mid-course did not assist with the acquisition of reliable results. Any future designs would incorporate custom H_2 or O_2 sensors developed at LANL. One yet to be determined result was an ever increasing signal corresponding to methane in the system. The repeatability and reliability of the RGA to detect this organic species is yet to be determined. Moreover, the purge gas was saved with the system post-irradiation, allowing for the potential subsequent analysis to test for the presence of CH_4 (section 5.3.).

As the system was calibrated with a H_2/O_2 mixture, the lack of a response for O_2, and the resultant assumption that the oxygen levels were below the detection limit, is also an issue that needs to be resolved. One possibility is that the silver metal in the iodine trap prior to gas removal for RGA analysis acted as a secondary catalyst for the hydrogen/oxygen recombination. This is indirectly supported by the fact the original calibration was done with the H_2 and O_2 mixture flowing through the system in a configuration that bypassed the iodine trap. Follow up studies with a bench top reactor could confirm or disprove this hypothesis.

3.3.2. Temperature Results

Plotted in Figure 5 are the temperatures recorded during the course of the irradiation for the reaction chamber, the outlet of the small heat exchanger following the recombiner, the tungsten target for neutron generation, and the Blue Room room temperature. The room temperature increased with the start of the beam, but then settled down on the second day. Similar trends were observed for the reaction chamber and heat exchanger as well. It was determined that this was caused by a small blower attached to the heat exchanger that failed. It was in fact heating the contents of the Lexan enclosure for the reactor and once it was turned off, the interior temperature dropped. The thermocouple to measure the room temperature was attached to the outside of the Lexan housing, so in fact the thermocouple was monitoring the wall temperature for the housing and not the room. The temperature for the tungsten target was more indicative of the state of the experiment and would fluxuate (increase and decrease) with the beam, and as would be expected the target would cool off dramatically when the beam was turned off.

3.3.3. Pressure Measurements

The Impress ceramic pressure sensors were very reliable and did not suffer performance issues during the irradiation. Their performance was the chief engineering safety metric for the irradiation. Presented in Figure 6 are the compiled pressure measurements internal to the reaction chamber. As shown in Figure 3 the MOP for the system was set at 0.25 bar (3.62 psig). This value was programmed into the control system for the experiment so that it that pressure was reached the beam would automatically be stopped, halting sample irradiation. Operators during the course of the experiment, however, also recorded the reactor pressure and paused beam when steep pressure increases were observed. This annotate plot (Figure 6) shows that after 27 h. of operation there was a rapid increase in pressure for the reactor and it was quickly decided to halt the experiment and turn off the beam to the target. Immediately a drop in pressure was observed. The experiment was suspended until an explanation for the event could be determined, or a remedy identified that could allow irradiation to continue safely. Analysis of the event over the following 24 h. determined that the most likely occurrence was a blocked flow through one of the components (tubing or iodine trap), with a less probable cause being a release of gas from the reaction solution and failure of the recombiner to adequately handle the increased hydrogen production. Once it was determined that the experiment could be safely operated by monitoring the internal pressure, and stopping the beam in the event of rapid pressure increases, the experiment proceeded at 49 h. Over the course of the next three days, various scenarios were tested with the experiment and it was found that increasing the flow through the pump would halt pressure increases and the internal pressure would return to normal. This in effect dealt with both possible scenarios, unblocking a clogged pipe (perhaps with water) or increasing the flow through the recombiner for a higher hydrogen turnover.

During the remainder of the irradiation, there was a gradual pressure decrease measured for the wider system. This was a result of continuous sampling to the RGA through the small leak valves and/ or possibly unexpected leaks into the purge tank or Lexan enclosure. However, no radiation above the expected background levels was detected for the 6 day irradiation. To compensate for the loss in pressure, argon was introduced to pressurize the system on multiple occasions.

After 118 hours of total irradiation time for the experiment it appears as though the diaphragm pump failed and the experiment was stopped, as control of the gas flow rate using the pump was the mechanism used to control pressure increases. Analysis of the system was planned to reveal the cause for this failure, either a materials failure or an unforeseen inherent mechanical collapse (See section 5.1.).

3.4. Conclusions

The gas handling team, working with the entire ^{99}Mo product effort, assembled a system to maintain, and recirculate, the overhead gas flow during the irradiation of the LEU solution within the LANSCE Blue Room. The system performed its function, allowing an LEU sulfate solution to be irradiated for multiple days without any loss of containment of volatile radioisotopes or build-up of potentially explosive concentrations of H_2 and O_2, the water radiolysis products. Monitoring various temperatures showed that the system remained near room temperature and adequate heat losses allowed for the recirculation of argon that would not heat or cause excessive evaporation of the LEU solution. Results from the RGA show that the stability of the instrument was not reliable; however, it appears as though the recombination of the radiolysis gases from water decomposition may have occurred in the iodine trap over the silver metal and not in the recombiner containing the platinum/palladium mixture. Lastly, it was demonstrated that vigilant monitoring of pressure changes within the system served as trustworthy metric for operating safely without over pressurizing the system. For any future irradiations it would still be prudent to equip them with H_2 sensing devices stable in the radiation fields to better measure the gas phase concentrations and stay well below the flammability limits for H_2 and O_2.

4. Irradiated Solution Sample Analysis and Separation Chemistry

4.1. Sample Retrieval, Gamma Spectroscopy Analysis and Titania Colum Separation Experiment at the CMR Hot Cells

4.1.1. Preparation of Titania Column Apparatus

The titania column separation apparatus was almost identical to the equipment used for the Target 4 ‘loop test’ separation chemistry experiments (Dec. ’12 – Jan. ’13). Separation equipment was composed of the following general components: source and collection vessels, tubing, column (*vide infra*), pumps, valves, and a heating block. The feed solutions and strip solution were held in plastic bottles, typically Falcon tubes. From these source vessels, 1/8” OD PTFE tubing led to the piston pumps (Eldex A-60-S) and 1/16” OD stainless steel (316) tubing led from the piston pumps to the column via valve heads. Swagelok pneumatic 3-way valves were used at the top and bottom of the column; valves were actuated only when the direction of solution flow was changed. The column was heated to 80 °C by surrounding it with a rectangular block of aluminum (4x4x12) with a cylindrical channel cut for the column and piping. The block was bisected vertically to allow it to be positioned around the column and piping. The block had adhesive silicone heating pads (BriskHeat) applied for hot cell operation. Thermocouples were used to monitor temperature, and a PID temperature controller (Digi-Sense) was used in conjunction with the thermocouples and heating block to control the temperature. A two-shelf stand was constructed to hold the equipment, and a spill tray was placed under the self.

0.6 x 5 cm stainless steel columns equipped with 2 micron frits at each column end (YMC America, XC0506WT) were packed with titania (Zirchrom Sachtopore-NP, 110 micron, 60 Å) that had previously been size-segregated using a micro sieve equipped with a 170 mesh (88 micron) screen; all particles that passed through the screen were discarded. The two columns were dry packed with ca. 1.9 g of TiO_2. Deionized water was passed through each column using a peristaltic pump for approximately 10 minutes at a flow rate of *ca.* 5 mL/min. The column was then submerged in deionized water in a capped bottle and sonicated for an hour. Using miniature, double shut-off quick-connect fittings (Swagelok), the columns could be attached and removed from the separation equipment. A new column was inserted into the assembly before each of the two separation experiments.

Prior to receipt of the irradiated sample from LANSCE, the separation apparatus was cold tested for leaks, flow rates, and dead (‘hold-up’) volume on the bench outside of the hot cell bank. The dead volume was determined to be 3.5 mL. Flow rates were set at 1.19 mL min^{-1} in the “up” direction and 1.39 mL min^{-1} in the “down” direction. After cold testing, the apparatus was installed in a hot cell and the piston pumps were appropriately primed using water. To ensure that no leaks had developed, the apparatus was again tested with water.

4.1.2 Sample Retrieval and Gamma Spectrscopy

When the drum containing the irradiated LEU solution was received at the CMR (March 5th, 2013), the outer drum was opened in front of a fume hood and a dose rate was measured at the lid of the inner drum. The inner drum was rigged and removed from the outer drum. At 30 cm, the dose rate was measured to be <5 mR/hour, and it was therefore stored in the Wing overnight. The following morning (March 6th), the inner drum was moved into the hot cell corridor. The lid was removed and dose rates were measured at contact inside the container (100 mR/hour) and at 30 cm (45 mR/hour). The fittings that connected the drain tube to the shipping vessel were too close to the lead shielding to remove by hand while the shipping vessel was still in place. Thus, personnel exited the hot cell corridor, and the manipulators were used to complete the unpacking process including detaching the drain tube and attaching a short, curved piece of stainless steel tubing to use as a pouring spout.

The shipping vessel was relocated from the hot cell corridor to a hot cell and the contents were decanted into two 175 mL Falcon tubes and one 15 mL conical bottom tube. The shipping vessel and the three tubes were weighed. Through weighing samples and the shipping container, and using the solution fuel density value of 1.192 g mL^{-1}, it was determined that 352 g (296 mL) of solution was recovered from the shipping container. Therefore 6.5 g (5.5 mL) of solution was lost as hold-up in the LANSCE apparatus after draining, in the shipping vessel after decanting and/or through water radiolysis/evaporation. 12.27 mL of solution was decanted into the 15 mL tube and this sample was set aside for subsequent analysis at TA48. In addition, this sample was analyzed directly by gamma spectroscopy at the CMR by Rawool-Sullivan et al. (LA-UR-13-23270). The 175 mL tubes contained 143.7 and 138.4 mL of solution, respectively, and were used for the titania separation chemistry experiments on two successive work days. By difference it was determined that < 1 mL of irradiated solution was lost during the three solution transfers.

4.1.3. Titania Column Separation Chemistry

Prior to beginning separation chemistry, the apparatus described in section 4.1.1. was heated to 80 °C for approximately one hour and the column was washed with 0.1 mol L^{-1} H_2SO_4 (Fisher) in the "up" direction. This solution was collected as the first fraction, and subsequently discarded. Next, an irradiated LEU solution was poured into the same source vessel and pumped through the apparatus in the "up" direction; it was collected as the second fraction. Subsequently, 1.0 mol L^{-1} H_2SO_4 (Ricca) and water were successively pumped through the apparatus in the "up" direction from the same source vessel; both were collected into the third fraction. Finally, 0.1 mol L^{-1} NaOH was pumped through the apparatus in the "down" direction to elute the Mo-99; and collected as the fourth fraction. Broadly, the VERSE simulation parameters provided by Argonne National Laboratory (developed in collaboration with Purdue University) were used for separation, as previously used for the Target 4 Loop test report (LA-UR-13-28967). For ease of operation in the hot cells, however, the specified flow rate for the uranium solution was used for all solutions flowing in the "up" direction. In addition, only three fractions were collected post separation; the uranium fraction, a wash fraction and a basic fraction that would hopefully contain all the Mo-99. In the previous Target 4 separation experiments 15 or 16 fractions were collected for analysis but, to simplify hot cell operations, only three fractions were collected for each of the two CMR titania column separations of Blue Room irradiated LEU sulfate solutions. For any future planed hot cell separation experiments additional fractions could be collected, if required.

During the second separation, the pump in the downward direction did not function properly. To finish the second separation, it was decided that all the solutions should be run in the "up" direction. Because of the high dose associated with the equipment it was impossible to determine the exact cause of the failure soon after completion of the experiment. From visual inspection and selective experimentation with water in the week after separation, one of two possibilities seems to explain the malfunction. (1) The PTFE tubing slipped out of its ferule and nut slightly, allowing the solution to flow back into the solution reservoir, thereby causing the pump to not be primed appropriately. (2) Precipitate or titania fines clogged the outlet tube. In the future, it may be appropriate to use a pump that is rigidly plumbed (possibly using 1/16" stainless tubing) at both the inlet and outlet to avoid unnecessary strain on the connection points. If required, a post-mortem could now be undertaken on this equipment as radioactive decay should be sufficient to allow detailed analysis.

Samples of the irradiated solution and the irradiated solutions post-column separations were shipped to TA48 for gamma spectroscopy, pH measurement and UV/Vis uranium concentration analysis on the 19th

March 2013. Samples of the base fraction and the two titania columns were shipped to TA48 for gamma spectroscopy analysis on 14th March 2013.

Pictures 6-11 illustrate the CMR hot cell operations.

4.2. Irradiation Solution Sample Analysis at TA48.

4.2.1. Target Solution Pre- and Post-Irradiation

Previous work at ANL indicated that peroxide, produced from radiolysis, could lead to uranium peroxide ($UO_4 \cdot xH_2O$) precipitation. In this case rather than radiolysis of 2 molar equivalents of H_2O resulting in the formation of 2 molar equivalents of H_2 and one molar equivalent of O_2 it would result in the formation of 1 molar equivalent of H_2 and 1 molar equivalent of H_2O_2. As a worst case scenario, all peroxide formation, it was calculated that 0.15 moles of H_2O_2 could be formed during sample irradiation, resulting in the precipitation of the majority of the 0.19 moles uranium present in the fuel (1:1 molar reaction). ANL have undertaken experiments that indicate that both Fe(II) and Fe(III) could catalytically decompose peroxide in uranium sulfate solutions, as also indicated previously in the literature. Therefore, to ensure that uranium peroxide precipitation was not an issue during sample irradiation in the Blue room the solution was spiked with Fe^{III} to 0.091 mM L^{-1} concentration in the fuel. Fe^{III} was used instead of Fe^{II} to eliminate the possibility of Fe^{II} reduction of Mo^{VI}, required for titania column separation, to Mo^{IV}. Post-irradiation there was no precipitation observed, and thus no evidence for formation of uranium peroxide.

On the 18th March 2013 three samples of irradiated fuel were returned from the CMR hot cells to TA48 for further analysis. The pH of the irradiated solution was recorded, pH 0.9, essentially unchanged from pre-irradiation (1.0), as was the density of the solution (1.19 g mL^{-1}). The fact that there has been no change in pH indicates no radiolysis of sulfuric acid, as would be expected for a comparatively 'small' radiation field (*vs.* plant operation) and comparatively 'high' sulfate radiolytic stability (*vs.* nitrate, the other potential target solution anion). Using the UV/Vis technique that we have previously developed, the uranium concentration was determined to be slightly higher post irradiation compared to pre-irradiation (156(1) gU L^{-1} *vs.* 150(1) gU L^{-1} pre-irradiation). The initial pre-irradiation concentration was also confirmed by Davis-Gray titration (150.3 gU L^{-1}, Target 4 report). This *ca.* 4 % increase in uranium concentration can in part be attributed to loss of water during irradiation (both radiolysis and evaporation), evaporation during hot cell operations at the CMR and additional systematic errors in the UV/Vis measurement not account for by the simple standard deviation error. A more detailed description of the application of this UV/Vis measurement technique was included in the accompanying Target 4 report (LA-UR-13-28967).

4.2.3. Radioisotope Production Values

On the 19th March 2013 irradiated uranium solution samples were submitted for gamma spectroscopy analysis following a 10 fold dilution in 0.1 mol L^{-1} H_2SO_4. Three samples in standard geometry scintillation vials were submitted for gamma spec analysis using a C-NR count room high purity germanium detector, and the results for ^{99}Mo, ^{103}Ru, ^{140}Ba, ^{141}Ce, ^{95}Zr, ^{147}Nd, ^{239}Np and ^{131}I production are shown in Tables 1-4 respectively (and Figure 7). Previously, Rawool-Sullivan *et al.* recorded the gamma spec of an irradiated solution sample through a port hole in the CMR hot cell (7th March 2013, LA-UR-13-23270) using a portable Ortec Transpec gamma ray spectrometer, and obtained production activities at End of Beam for ^{99}Mo, ^{103}Ru and ^{140}Ba.

Using the 181 keV ^{99}Mo transition a production value of 67.70(3.75) mCi was obtained using the Transpec instrument vs. 68(8) mCi by the count room measurement. This would appear to be very good agreement, apart from the high error associated with the count room measurement (partly due to one particularly high count reading for one sample). Comparing the activities obtained using the 740 keV transition, 67.74(3.73) mCi - Transpec *vs.* 58(1) mCi – count room, there is more of a difference between analyses. While gamma spectroscopy measurements using a well calibrated high purity germanium detector are usually very accurate, the count room measurements were made 17 days after end of beam and long count times were required to obtain acceptable counting statistics. In addition, there could perhaps be a more detailed investigation into removal of any contribution from the nearby ^{95}Zr transition (at 757 keV), a dominant transition 17 days after EOB, from peak analysis of the 740 keV ^{99}Mo transition. In conclusion, as three of the four sets of gamma spectroscopy analysis indicate that 68 mCi of ^{99}Mo were produced at EOB of the Blue room irradiation then this is the most likely experimentally measured production value, which is in good agreement with the model value which projected that 65 mCi would have been produced at EOB based on beam history.

Less than 1% count time errors were obtained using the count room gamma spectroscopy analysis for ^{103}Ru and ^{140}Ba, both longer half-life radioisotopes than ^{99}Mo, yielding end of beam production activities of 3.26(1) mCi and 18.32(5) mCi respectively. These values are *ca.* 10 % lower than reported using the Ortec Transpec gamma ray spectrometer, 3.665(0.190) mCi and 20.47(1.07) mCi, although in each case the values are not significantly different when taking into consideration measurement errors. In addition, production activities could also be obtained for ^{141}Ce, ^{95}Zr, ^{147}Nd, ^{239}Np and ^{131}I, although in the case of ^{131}I this only relates to the non-volatile fraction that remained in solution.

4.3.3. Separation Chemistry Results

As described previously, 129, 128 & 136 mL of irradiated Target LEU solutions were fed through the titania column separations for the loop test Target 4 experiments with near quantitative recovery of ^{99}Mo in the base fractions (LA-UR-13-28967). Numerous column fractions were collected and analyzed to provide detailed analysis of the column separation chemistry of many key radioisotopes. In contrast, as this was the first hot cell operation performed with the separation equipment it was decided to simplify the operating procedure by eliminate changes in flow rates when switching between feed and wash solutions. In addition, the number of collected column fractions was greatly reduced from 16-17 to 4: - (i) The 0.1 mol L^{-1} H_2SO_4 wash (not analyzed), (ii) the uranium solution post titania contact, (iii) the water and acid washes combined (not analyzed). and (iv) the 0.1 mol L^{-1} NaOH base strip solution. Feed solution masses (and volumes) and collected fraction masses (and volume, where available) are shown in Tables 5 & 6. The first separation experiment was undertaken on the 11th March 2013 and the second on the 12th March. 2013, with column end times of 70.9271 and 71.8507 respectively and all gamma spectroscopy data calculated to column end. The first column proceeding as expected. During preparation work for the second separation experiment the piston pump used for the 'down' delivery of

0.1 mol L^{-1} NaOH was inoperable and thus all the feed solutions, including the NaOH strip solution, were fed through the column in the 'up' direction (see section 4.1.3). Only the uranium feed solution and NaOH base strip solutions were analyzed quantitatively by gamma spectroscopy, samples arriving 18th and 14th March 2013 respectively. In addition the two YMC America stainless steel columns arrived on the 14th March. The dose rates on these two columns were too high to consider removing the post-column titania for direct measurement, but the two samples were contained in HDPE bottles and non-quantitative gamma spec analysis performed.

The solution density, pH and uranium concentrations of the uranium solution post column separation were obtained. The density values were identical, 1.18 g mL^{-1}, while the pH values were near equivalent, 1.0 for the first separation experiment sample and 0.9 for the second separation experiment sample. The uranium concentrations were also effectively equivalent, 151(1) g(U) L^{-1} for the first separation sample and 150(1) g(U) L^{-1} for second. The slightly lower post-columns uranium concentrations vs. the feed solution (156(1) g(U) L^{-1}) is probably partly due to slight sample dilution through co-collection of a fraction of the 0.1 mol L^{-1} H_2SO_4 wash solutions present prior to passing the irradiation uranium solutions through their respective titania columns. As expected from previous studies, ^{140}Ba, ^{141}Ce and ^{147}Nd pass through the titania column with the uranium fraction with good activity balance (Tables 7-9). It would also be expected that ^{239}Np would not binding to titania, but in this case the activity balance is not as good (Table 10). This is probably a result of the higher count errors due to the short ^{239}Np half-life and the long time delay between EOB and analysis. In both column experiments *ca.* 90 % of the ^{105}Ru passed through the column with the uranium product (Table 11). In the previous Target 4 experiments significantly less (*ca.* 60 %) of the ^{103}Ru passed through the column with the uranium product, a major fraction remained irreversibly bound to titania and a small, but significant and measureable, fraction was eluted with the ^{99}Mo product in the base solution. This may suggest a more reducing environment where there is more Ru^{III} present. The other trivalent cations radioisotopes that we have analyzed do not bind to titania e.g. Ln^{III}, $^{105}Rh^{III}$. As a consequence of the presence of more Ru^{III} there would be less Ru^{IV} present which would probably bind to titania, all other tetravalent cations binding irreversibly to the column *e.g.* $^{95}Zr^{IV}$. At present this is just a working hypothesis, a more detailed understanding of tracer ruthenium chemistry in irradiated uranium sulfate solutions would be required to substantiate this hypothesis.

Using the 181 keV transition the % recovery of ^{99}Mo in the base fractions were 85 and 80 % for the first and second separation, respectively (Table 12). Following the 740 keV transition the % recovery of ^{99}Mo in the base fractions were 95 and 90 % for the first and second separation experiments, respectively (Table 13). In each case more ^{99}Mo is recovered in the base fractions in the 1st vs. the 2nd column separation experiments. It would appear that the pump malfunction in the second column separation, restricting all solution flows in the 'up' direction of the column, had a measureable impact on ^{99}Mo recovery. Presumably most the ^{99}Mo is bound to the bottom of the column so when the flow direction can be changed to the down direction ^{99}Mo can be efficiently eluted (as anticipated by the ANL column design). The discrepancy observed in ^{99}Mo recovery using the too different gamma energies can be attributed to the 'low' production value obtained for ^{99}Mo using the 740 keV peak, which would point to a maximum 85 % recovery of the medical isotope in the base fraction. *i.e.* the 180 keV data is probably the more reliable. This would also tie in with the gamma spectroscopy data on the two columns where transitions attributed to ^{99}Mo were clearly observed in both cases, although quantitative analysis was not possible. In analogy with the ^{103}Ru data, this points to partial reduction of Mo^{VI} to Mo^{IV} which then irreversibly binds to titania. However, what is not known is whether Mo^{VI} reduction occurred during sample irradiation in the Blue room or in the 9+ days between EOB and the first column separation.

As expected, the ^{131}I is split between the irradiated uranium solution and the ^{99}Mo base fraction post separations chemistry (Tables 14 & 15), with ^{131}I also observed bound to the titania column and presumably in the water/1.0 mol L^{-1} H_2SO_4 wash fractions (at least for the second column experiment). Interestingly, significantly more ^{131}I is recovered in the base fraction of the 1st separation than in the 2nd separation providing more evidence that passing the 0.1 mol L^{-1} NaOH base in the down direction (separation 1) *vs.* the up direction (separation 2) significantly increases efficiency of stripping radioisotopes from the column. It is also worth noting that in column experiment one there is effectively quantitative recovery of iodine when combining recovery in both the base and post-column uranium solutions, indicative of no loss of volatile iodine.

5 Blue Room Apparatus Post-mortem

5.1. Decommissioning the Apparatus

After the 26th Feb. – 2nd Mar. 2013 Blue room irradiation and irradiated uranium solution and cooling water drain-downs, the reaction apparatus was rolled to the side of the Blue Room and allowed to decay store for several months. In July - August 2013 the apparatus was secured, surveyed, and transported to the Actinide Research Facility (ARF) at TA48 where it was dismantled for post-mortem analysis. The Residual Gas Analyzer (RGA) and Pump carts were also moved to TA48; the pump cart was disassembled and the components were bagged and stored in the ARF. The scroll pump from the RGA cart was removed, bagged, and stored, while the RGA and turbo pump were put into service in the ARF.

5.1.1 Lessons learned during the decommissioning process

Decommissioning the apparatus was successful, with low exposure to radiation fields and only minor radioactive contamination observed on components during disconnects in which contamination was expected. In hindsight it is surprising that so few problems were encountered during decommissioning considering the unique nature of the project. However, during this process a number of observations were made, lessons that can be applied to any future undertaking of this nature.

1. Ensure that potential problems associated with decommissioning are extensively explored at the beginning of the project, during procurement, and during construction of complex apparatus. Several problems appeared in the dismantling process that could potentially have been avoided with more rigorous front end planning. For example, during dismantling several sharp corners were exposed which introduced cut hazards.
2. Consider how the behavior of a cart will change with several hundred pounds placed on it, much of the weight due to the lead shielding. In this instance it became more difficult to handle and having four steerable wheels would have made it more maneuverable. However, the cart was not so

difficult to maneuver that it couldn't be transferred between two sites several miles apart at LANL, and between different buildings on the same site.

3. As far as possible, match all components in terms of quality and double- and triple-check system components in terms of material compatibility. In the Blue Room experiment the only vital component that failed, the Viton circulation pump, lasted until almost the very end of the allotted time for the experiment.
4. The expertise for constructing the Blue Room apparatus rested with scientists/engineers who had limited experience with working with radioactive materials, and thus a team on the ^{99}Mo project with rad-work experience had to be assembled to undertake the decommissioning. To overcome the lack of detailed knowledge possessed by the team decommissioning the apparatus, the workers who were instrumental in building the apparatus were present to give advice, guidance and supply vital tools and equipment. This combination of expertise worked very well, but it was essential that all the workers with the required range of expertise could make time available to complete the task.

5.1.2. Decommissioning steps.

In general, the reaction chamber skid was dismantled using the following rationale:

1. Vent entire reaction system, except the Holding/Purge Tank.
2. Remove Lexan sheets as necessary to remove interior components.
3. Remove the lead shielding from around the Purge Tank.
4. Remove the LEU target solution shipping cylinder (used to ship the original LEU target solution from TA48).
5. Remove the Reaction Chamber.
6. Remove the Iodine Trap.
7. Collect gas sample from Purge tank.
8. Vent and remove the Purge tank.

The work was conducted under an appropriate Integrated Work Documentation and Radiological Work Permit. At each disconnection point, the joints were surveyed and/or swiped to look for contamination. The most significant dismantling steps are outlined below

A. Reaction system venting. A ¼" plastic line was connected at the appropriate valve and run into a fume hood. The fume hood-end of the line was lowered into a water-filled beaker to look for the evolution of gas bubbles. There was a very slight possibility of higher-than-ambient pressure in the system, although the reaction was kept at 2 psig for the duration of the experiment. The valve was opened, with 60 psi argon to the actuator. Only 7-10 bubbles were observed over a 15 min period, indicating that there was no pressure buildup.
B. The next steps involved removing all the components with potential contamination of irradiated LEU solution or fission products. 830 mg of uranium still remained unaccounted for, and was presumably still in the transfer vessel and reaction vessel (+ associated piping) associated with the Blue room apparatus, with some also remaining in the transfer vessel shipped to the CMR. Attempts to recover this material were required for accountancy purposes. Thus the containers that could contain LEU

were disconnected and transferred into a fume hood for further manipulations. Trace levels of contamination were observed during these operations.

C. The iodine trap was removed by separating the VCR fittings at each end of the trap. Both fittings were bagged; a smear of the top fitting showed minor contamination and was capped immediately. The bottom fitting showed no contamination and was also capped. The iodine trap was then analyzed by gamma spectroscopy, in a secondary container, with no detectable activity observed. This is not surprising as during the months between EOB and final decommissioning any ^{131}I present would have decayed to tracer levels.

D. With the major components removed from the front of the reaction chamber skid, the disassembly moved to the back of the apparatus. The 5 gallon purge tank required venting, but a sample was taken before venting for analysis of the mixture. An evacuated sample bottle was attached to the system and a sample of purge gas collected. The purge tank was then vented by attaching a ¼" plastic line, with the line lowered into a water filled beaker in a fume hood. After carefully opening the appropriate valve to the Purge tank copious gas evolution was observed, as would be expected from purging a 5 gal. volume tank at 2 psig pressure. The apparatus was left in an "open" configuration to ensure complete venting over a period of 4 days. The tank was then purged with argon for 5 min. at just above atmospheric pressure.

E. The gas sampling manifold (leak valves and automatic valves) were removed, including the gas sample itself. The leak valves were not contaminated, and were bagged and saved in case required for later experiments.

F. The rupture disk and the lines around the rupture disk were removed as a single unit. To help determine whether a leak in the rupture disk could in anyway have caused the observed pressure transients during irradiation it was further analyzed, but there was no evidence for degradation (see also section 5.4.).

G. The purge tank was removed by unfastening the nuts/bolts on the bottom flanges, and prepared for disposal.

H. The recirculation diaphragm pump was removed and placed in a fume hood for later examination. There was no evidence of mechanical seizure or any problems with the electrical connections within the pump and thus it is still unknown why this pump failed towards the end of the sample irradiation

I. The junction box on the back of the cart was removed, as were the remaining Lexan panels and the 80/20 aluminum frame – thus completing the decommissioning.

5.2. Irradiated Uranium Fuel Accountancy

As stated in section 4.1.2. , 5.5 mL (6.5 g) of LEU solution remained unaccounted for after transfer of the irradiated solution to the CMR in Mar. '13. While this amounted to only 2 % of the total solution volume it still represented an accountable quantity of uranium (830 mg) and thus the LEU solutions remaining in the two transfer vessels (TA48 to LANSCE and LANSCE to CMR) as well as the reactor vessel all had to be washed out with water to recover the uranium. In each case the recovered uranium solutions were still yellow, indicating that even after sitting in stainless steel vessels for months there was limited corrosion (analysis of corrosion in these systems is described in detail in the associated Target 4 report, LA-UR-13-28967). After reducing the solution volumes and analysis by UV/Vis spectroscopy the quantities of uranium found in the TA48 to LANSCE transfer vessel, LANSCE to CMR transfer vessel and the Reaction Chamber were 102 mg, 173 mg and 199 mg respectively. Thus, in total, <400 mg of uranium remained unaccounted for, a sub-accountable amount according to DOE regulations which state that enriched uranium should be accounted for down to the 0.5 g level. In total >99 % of the total uranium was recovered post-Blue Room irradiation experiment.

5.3. Purge Tank Analysis

The procedures relating to undertaking gas analysis were documented in detail in the associated Target 4 report. Connecting the gas sample bottle (300 mL volume) to the purge tank left *ca.* 10 mL of manifold between the two that was not evacuated. From subsequent analysis of the gas pressure in the bottle, and assuming atmospheric pressure in the manifold, it was calculated that 97 % of the gas in the sample came from the purge tank and 3 % from air. Using an RGA (Residual Gas Analyzer), and a calibrated air sample (for % O_2 and % N_2), it could determine that *ca.* 55 % of the gas in the sample was air, with the balance being argon. In some respects this is not surprising as the purge tank was not 'purged' of air prior to opening to the gas handling system (containing mainly argon) at the end of the experiment. Interestingly, using the RGA analysis of either O_2 or N_2 a value of 55% of air in the purge tank was obtained. This indicates that little, if any, radiolysis produced O_2 is present in the purge tank, which in turn indicates that the O_2/H_2 recombiner (or the iodine trap) was effective in scrubbing out oxygen.

5.4. Leak rate analysis of a rupture disk

5.4.1. Background

During the irradiation process at the Blue Room it was noted that there were periodic pressure cycles, when the irradiation was started, where the pressure would initially increase followed by slow decay. Theoretical calculation of gas generation predicted increase in pressure due to radiolysis, Figure 4. However a steady state should develop where the recombination would counter continued pressure increase. This was assumed to be when the oxygen concentration begins to increase at approximately 10 min. or 2.4 psig.

The system was leak check multiple times before the experiment which revealed no leaks in the system. During discussions of the pressure transients, it was hypothesized that the pressure safety rupture disk may have had hydrogen diffusion across the graphite disk which might have caused the pressure decrease.

As part of the post-mortem analysis, a test system was assembled to determine potential hydrogen diffusion across the graphite rupture disk. A rupture disk module was used that was identical to the rupture disk on the Blue room apparatus. The rupture disk was considered potentially contaminated and it was not possible to release it for post-test at the cold laboratory facilities equipped for such testing, only visual confirmation of no obvious degradation (section 5.2.) The 'cold' rupture disk used for this cold testing was acquired from Zook. The disk was 1.5” diameter, constructed of *MONO* Graphite, with a burst pressure of 7 PSIG.

It was noted that when the Blue Room gas handling system was disassembled the system was still under pressure (above atmospheric pressure) indicating that the rupture disk seals did not have a leak through the rubber gasket flange assembly. The rupture disk and assembly are shown in Figure 8.

5.4.2. Test Setup and Method

The test system setup is shown in Figure 9. To determine if there was any diffusion through the graphite disk, the disk was pressurized with ~2 psig surrogate gas consisting of 4% hydrogen in a nitrogen balance. The system was then isolated from the source. Diffusion across the graphite disk would show pressure increase on CH2 with a decrease in CH1. If CH1 decreased, and CH2 did not increase, this would be considered as leakage through the rupture disk flange assembly, and not through the graphite disk. Pressure decay recordings were made periodically (roughly every 1 minute) after the pressure was isolated from the source. The resultant leak rate was recorded in column "Leak Rate" in Tables 16 and 17.

Between tests 3 and 4 it was discovered that the original torque value used on the bolts (5 ft-lbs) was the incorrect torque value. The proper torque that was supposed to be used was 17 ft-lbs. As such, tests 4-6 were performed with the bolts tighten to a torque value of 17 ft-lbs to see if the leak rate was different from the first two tests.

A Restek leak detector was used to determine the area around the rupture where the leaks occurred. This leak detector does not indicate leak rate. Instead, it sounds an alarm and LED lights illuminate when leak has been detected. Figure 10 shows the regions where leaks were detected. The leaks that were detected between bolts 1 and 4, and, 3 and 4 were discovered only during the first three tests. All subsequent tests showed leaks only between bolts 3 and 4. The following data shows the leak rates that were recorded. It should be noted that the first test (Test 1) has inconclusive data. However, the data has been included in the report as information.

From data (decrease in CH1 and no increase in pressure on CH2) and leak detector results, a leak at the flange gasket was identified. CH2 readings showed little change and could be attributed to temperature fluctuations and pressure detector drift. However, the readings from CH1 definitely show leakage (pressure drop), which appear to be leakage through the flange seals of the rupture disk assembly, as noted by use of the Restek leak detector.

5.4.3. Conclusion

Considering that there was still pressure on the Blue Room apparatus prior to disassembly, the rupture disk on the Blue Room apparatus did not leak in the same way that the test rupture disk leaked. Further, this would imply that the pressure fluctuations seen during the radiation experiment in the Blue Room were not related to leakage from the rupture disk. In addition, the leak rate seen in the above experiments were much greater than those seen in the Blue room irradiation experiment, estimated to be 0.04 torr/min. Therefore, leakage through the rupture disk was likely not the cause of the pressure fluctuations observed during the Blue Room irradiation.

Pictures

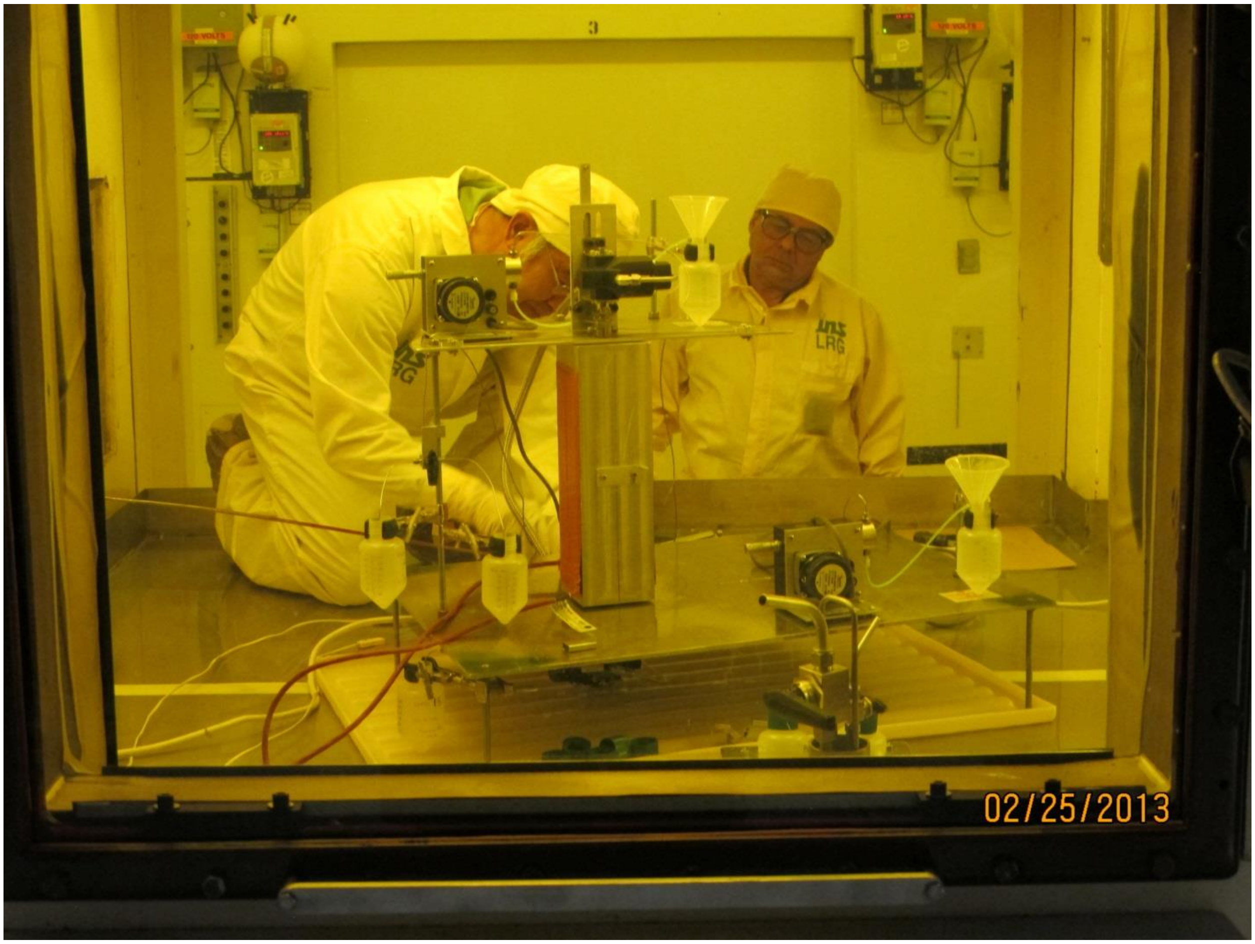
Picture 1. Blue Room Apparatus for the LEU solution irradiation

Picture 2. Ionex Ag-exchanged zeolite.

Picture 3. Air Dimensions Inc., single head B-series Dia-Vac pump.

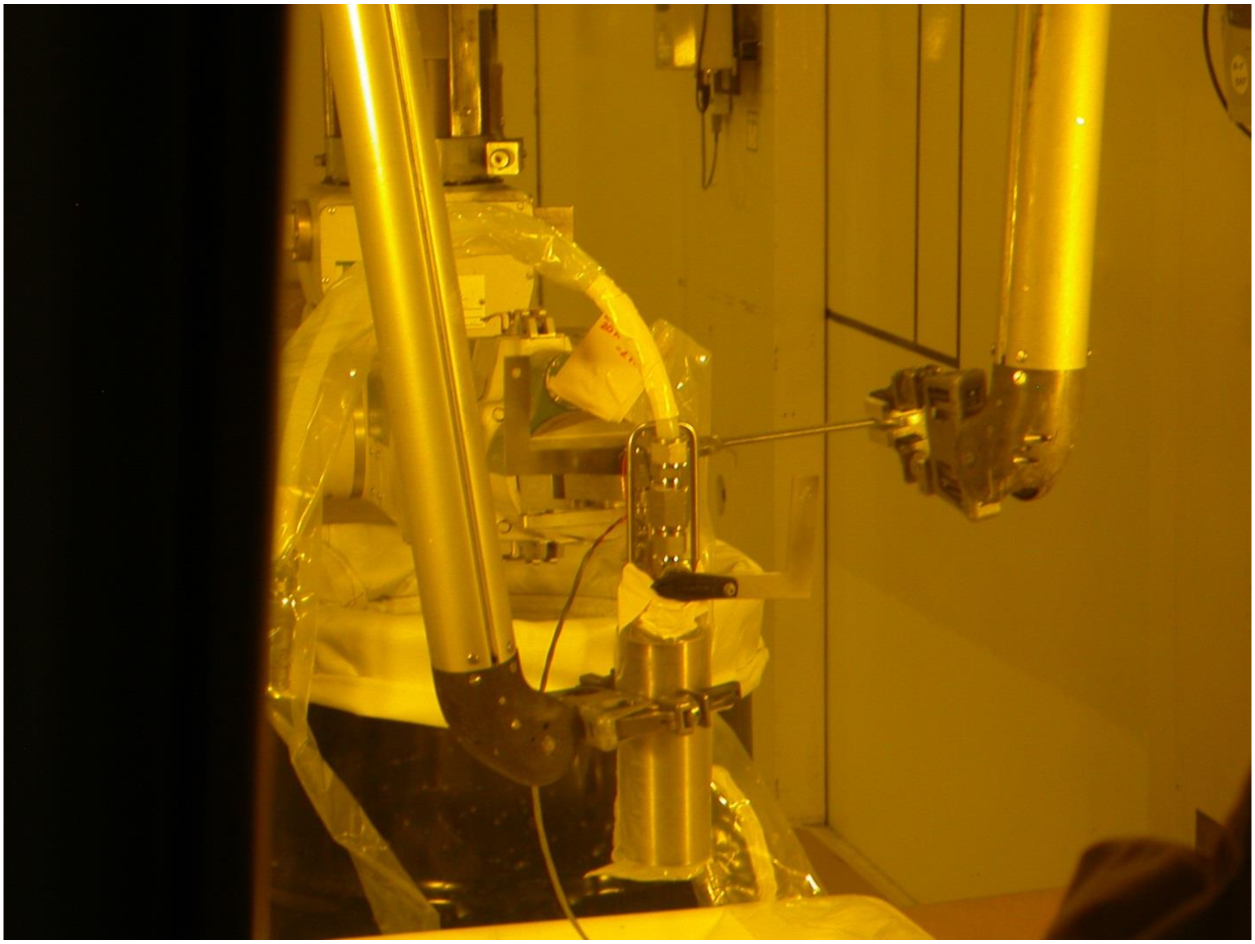
Picture 4. H_2/O_2 Recombiner from Resource Systems, Inc. The approx. length is 8".

Picture 5. SRS RGA-100 mass spectrometer.

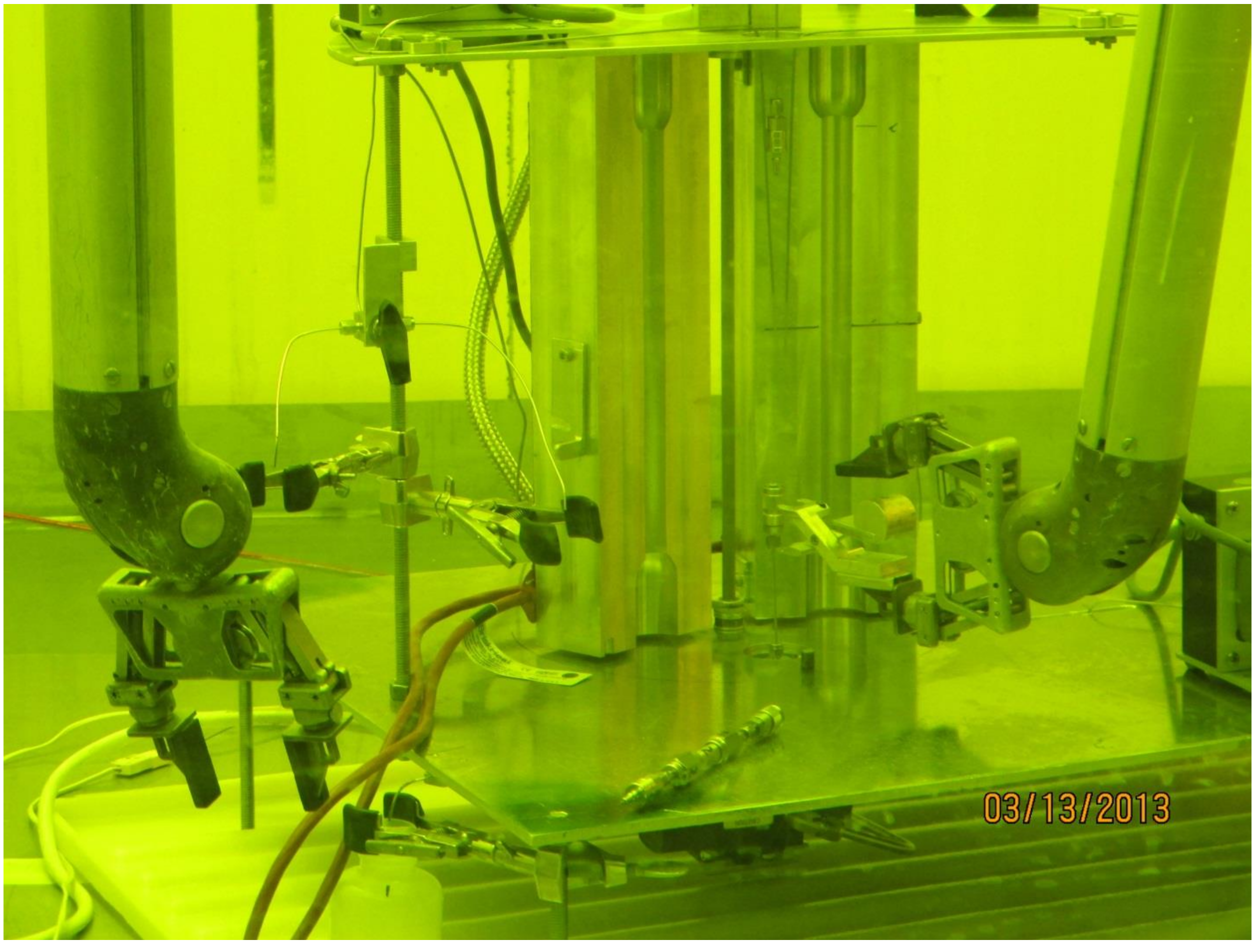


Picture 6. Installing the separation apparatus in the hot cell.

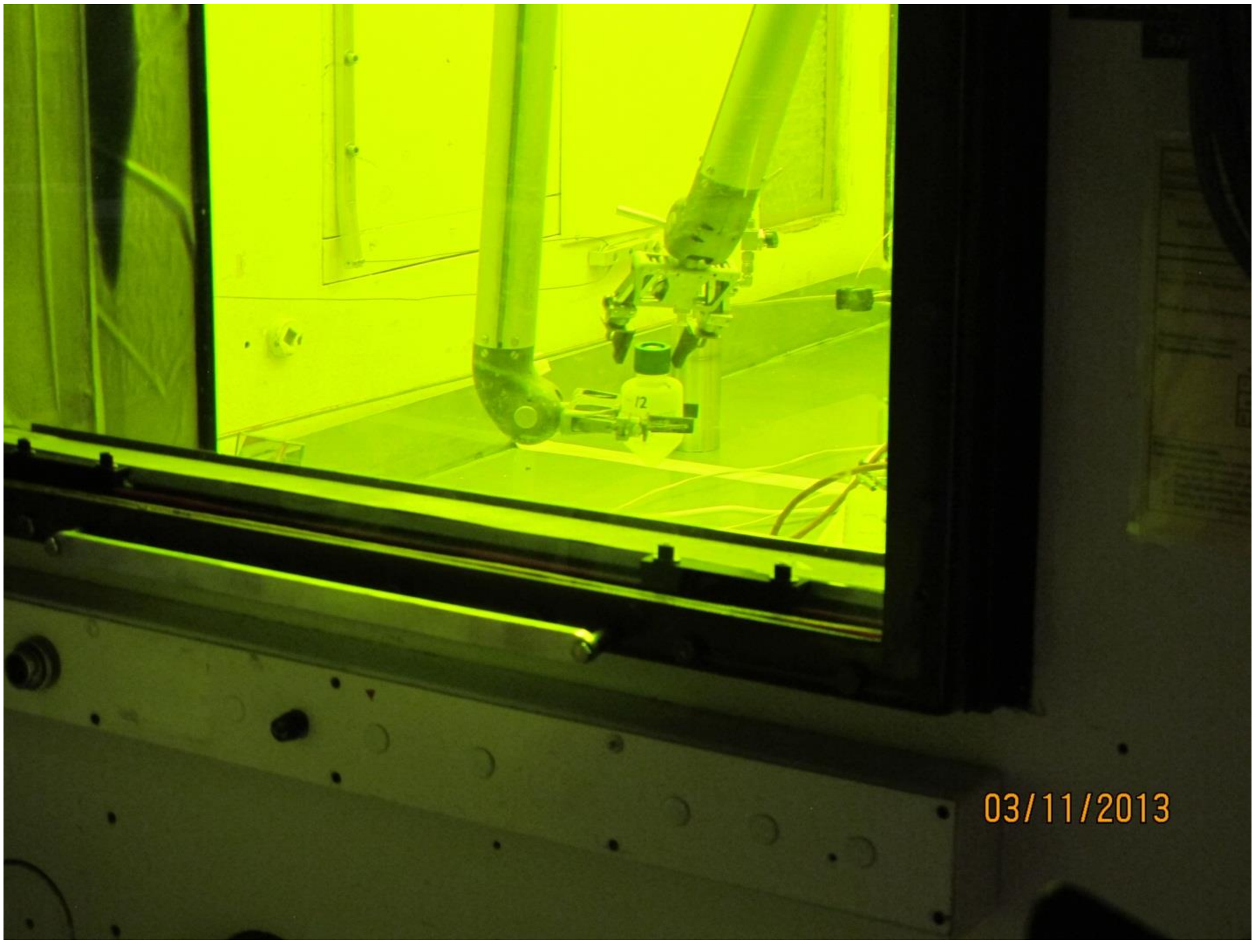
Picture 7. Lifting the inner drum out of the outer 9979 shipping drum.



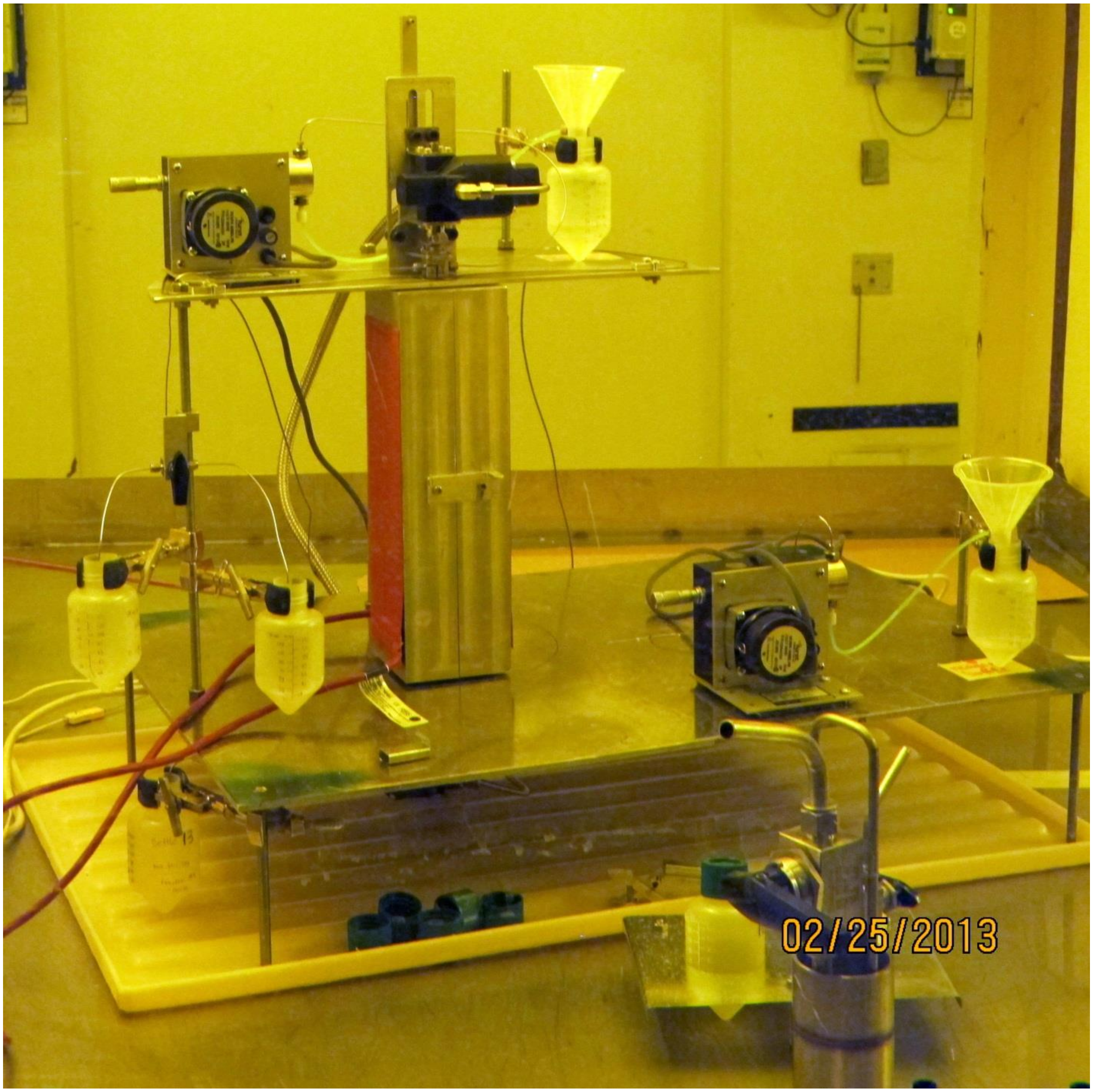
Picture 8. Using manipulator hands and a wrench to detach the drain tube from the shipping vessel in the hot cell corridor.



Picture 9. 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).



Picture 10. Using the manipulators to uncap a bottle.



Picture 11. Separation apparatus in the CMR hot cell.

Figures

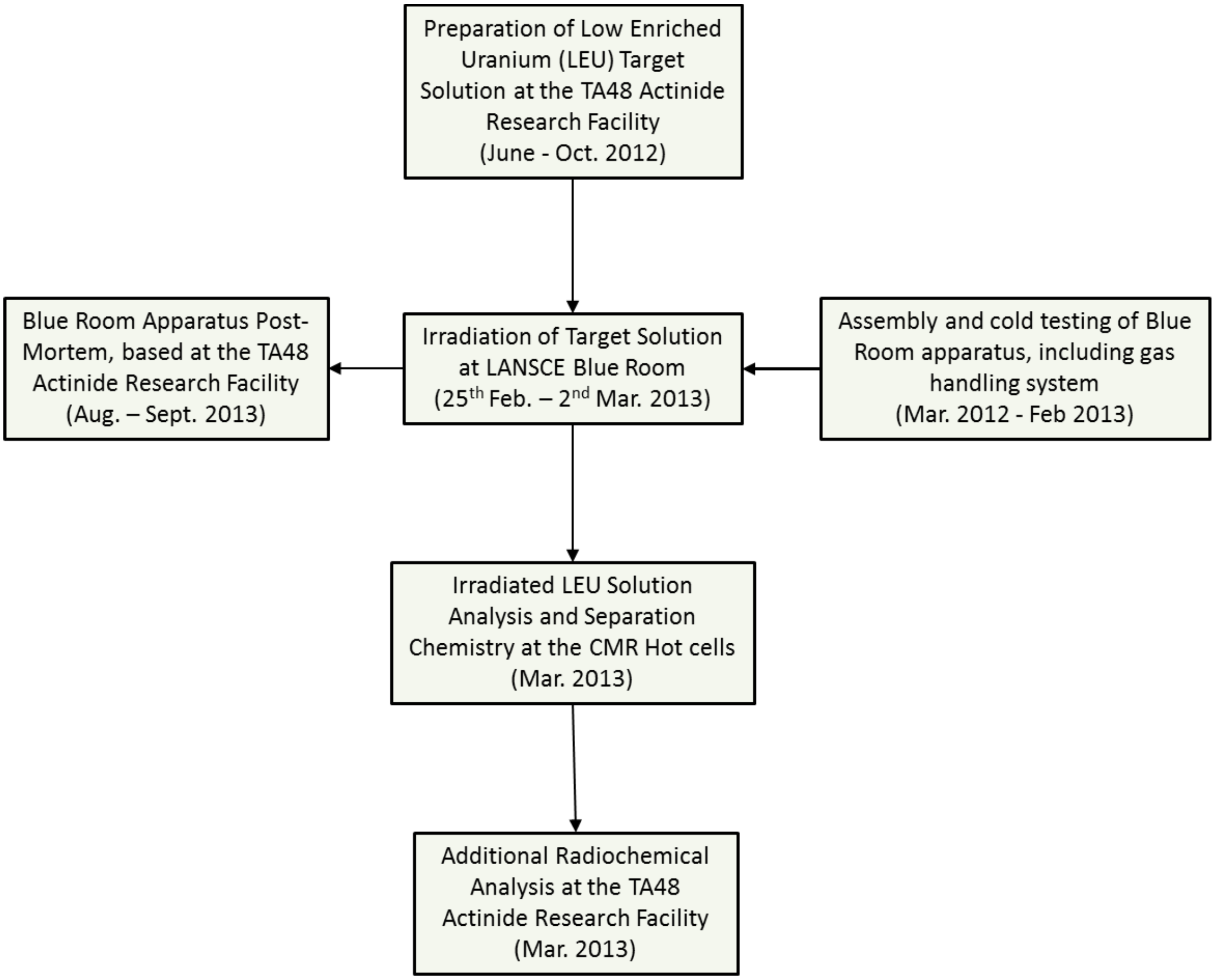


Figure 1. Schematic showing the Major Technical Steps undertaken for the FY 2013 Blue Room experiment.

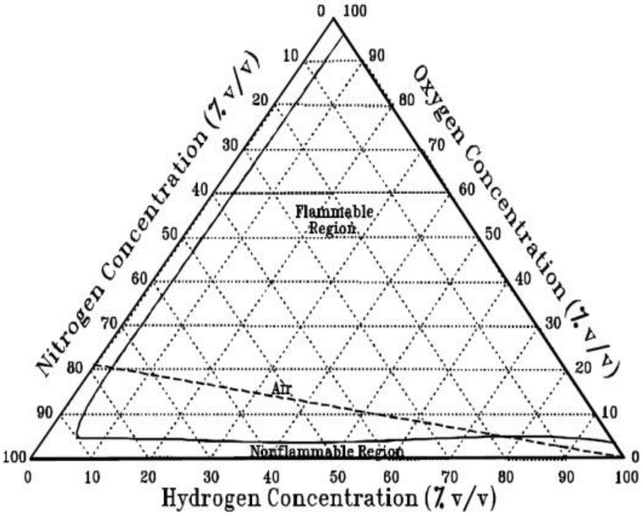


Figure 2. Flammability diagram for H_2, O_2, and N_2 (NASA, NSS 1740.16, Safety Standard for Hydrogen and Hydrogen Systems).

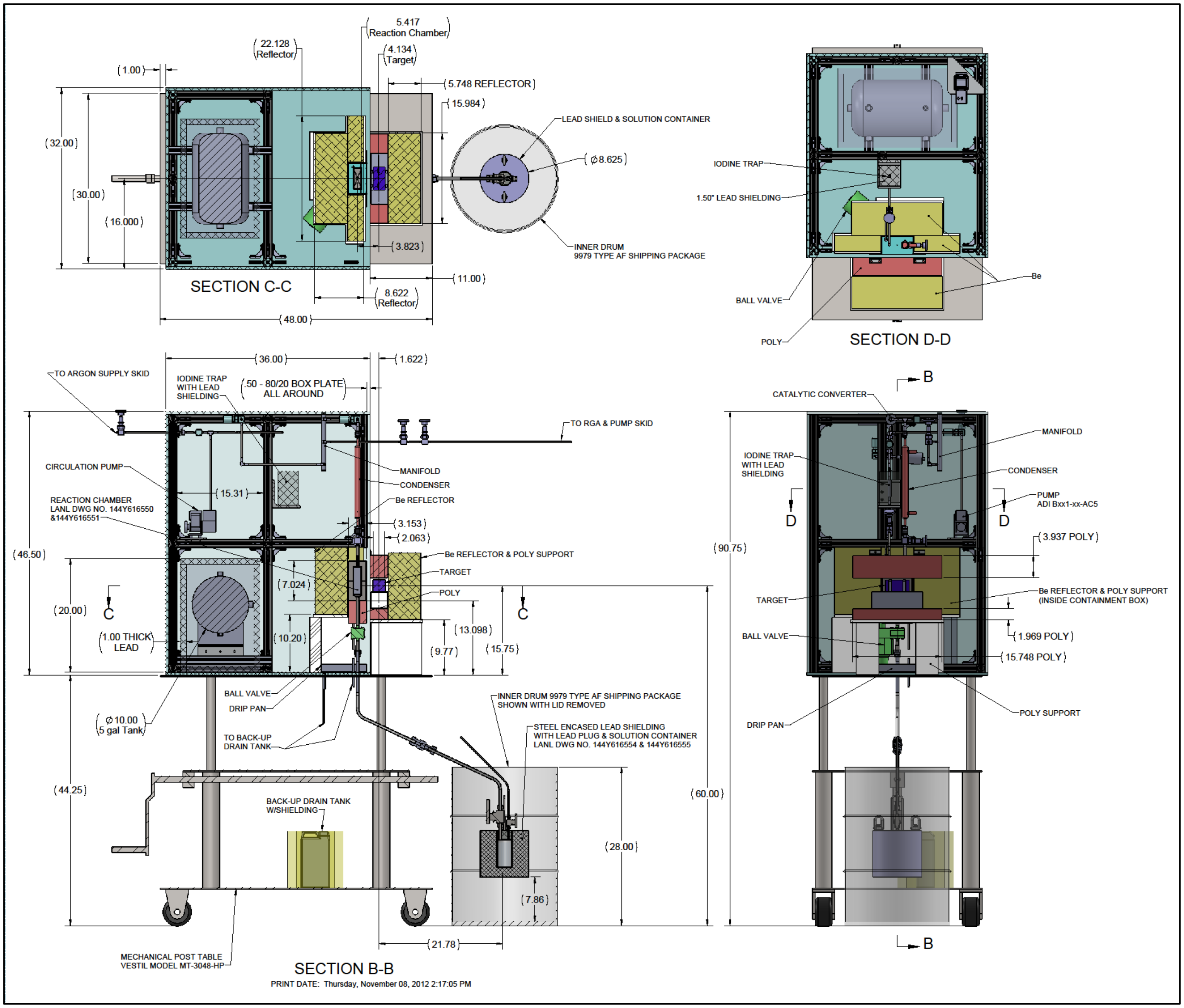


Figure 3. Drawings of the Blue Room Apparatus for the LEU solution irradiation

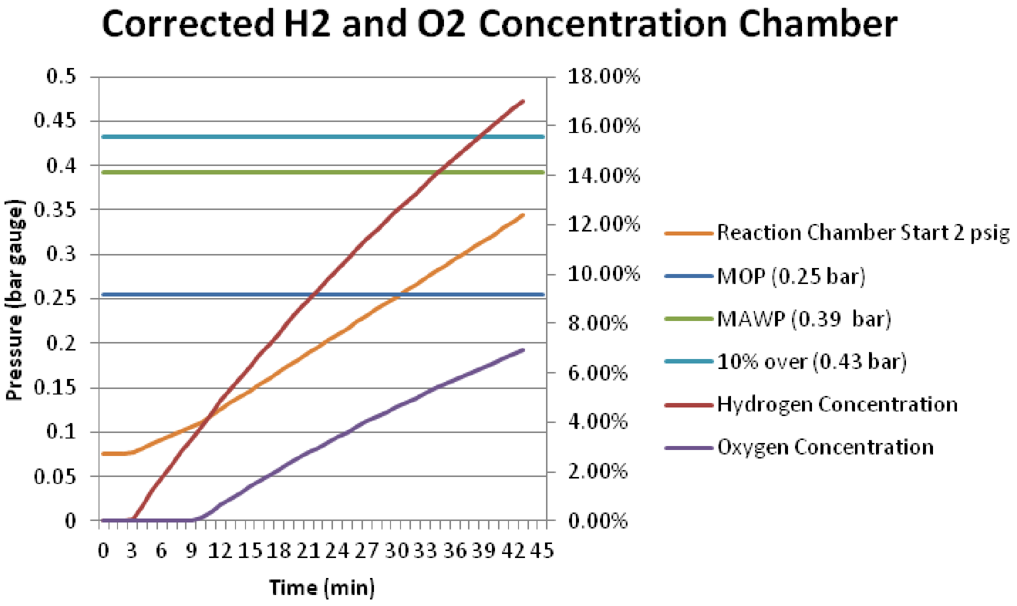


Figure 4. Pressure and concentration increases with time during operation of the gas handling system if assuming the absence of a catalytic H_2/O_2 recombiner (*i.e.* if the catalytic converter fails during operation). MOP is the maximum operating pressure for the reaction system, as determined by the research team. MAWP is the maximum allowable working pressure determined by the reactor design and Pressure Safety requirements at LANL. (1 psig = 0.069 bar).

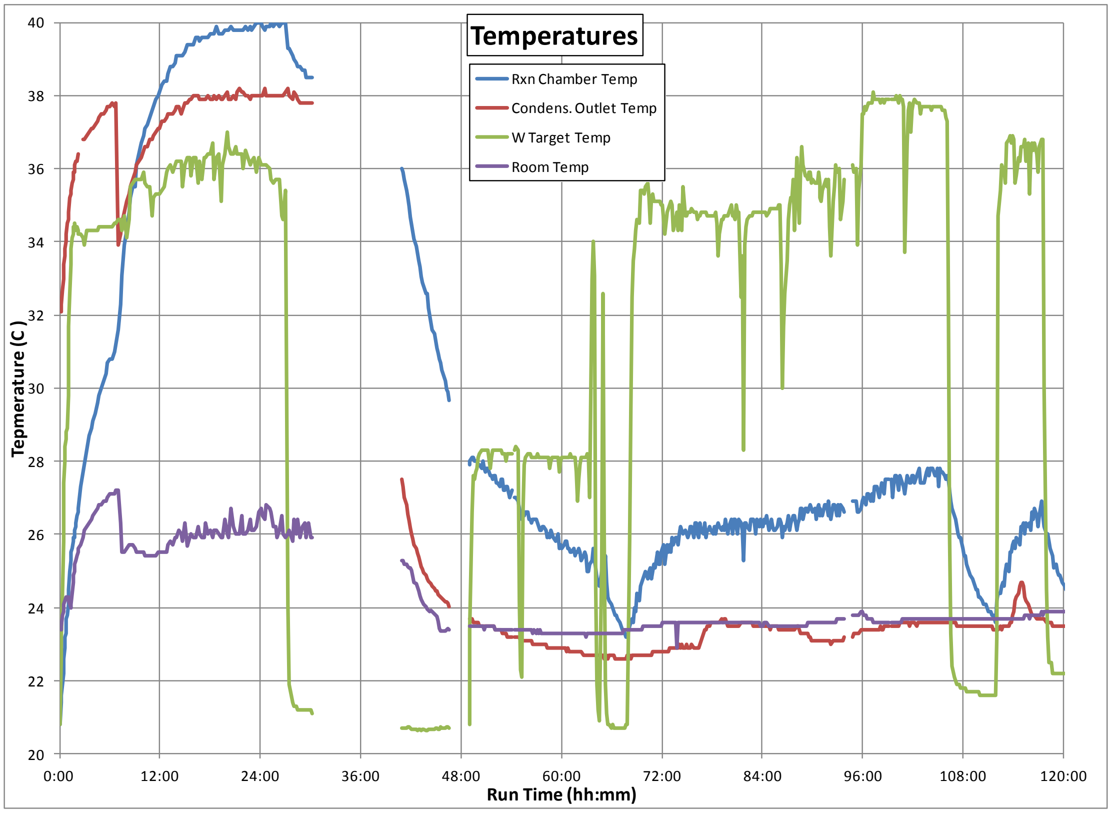


Figure 5. Recorded temperatures for the different components/areas during the LEU target irradiation.

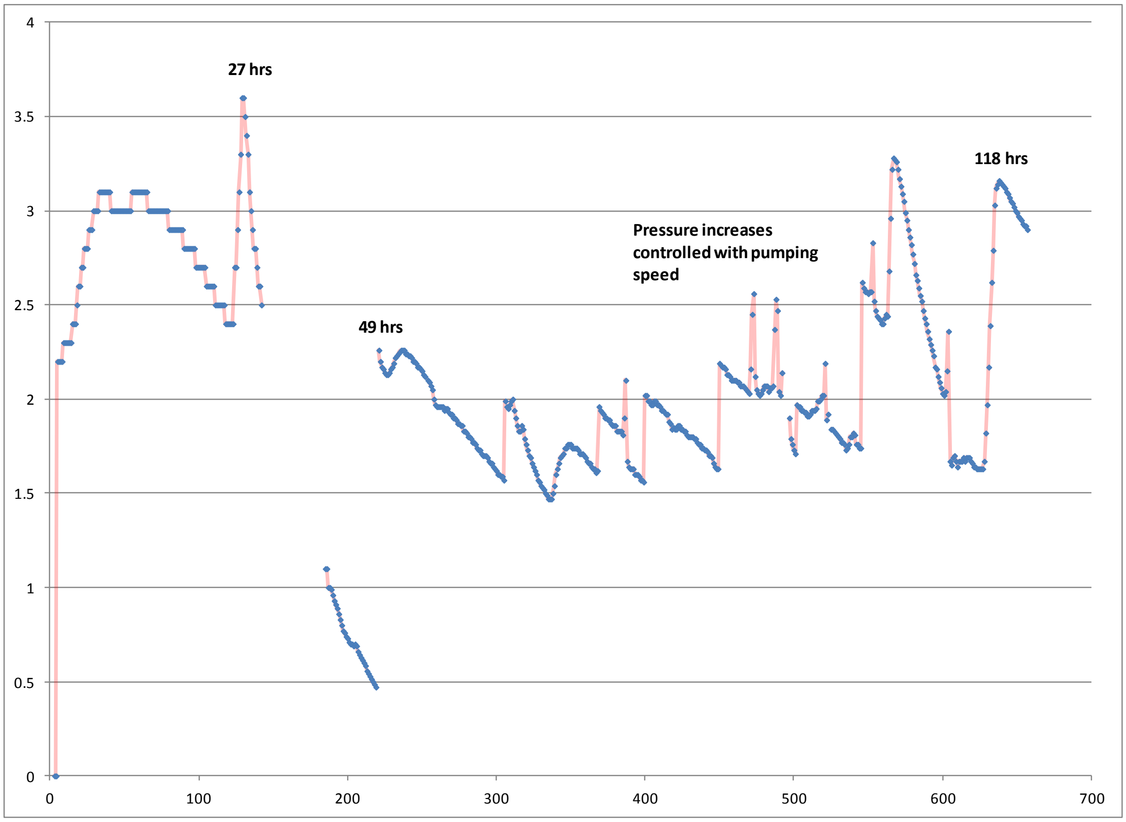


Figure 6. Recorded pressure values for the reaction vessel in psig (y-axis), recorded versus time (arbitrary units, x-axis).

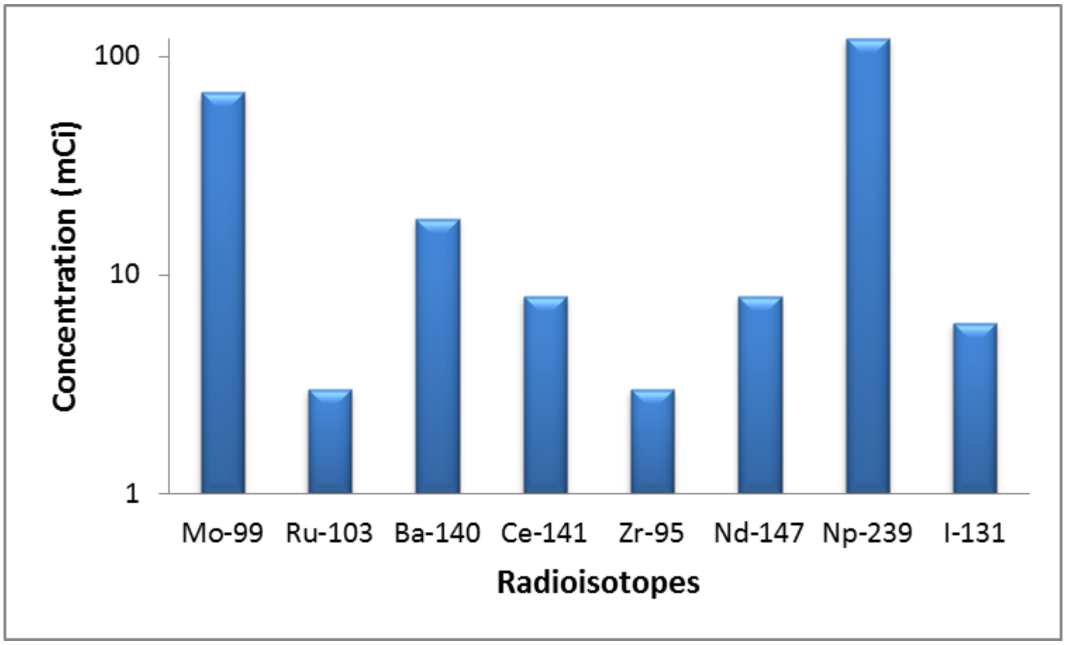


Figure 7. Production values (mCi) of main radioisotopes generated in the Blue room irradiated LEU sulfate solution. Gamma spectra recorded *ca.* 18 days after EOB (61.9167).

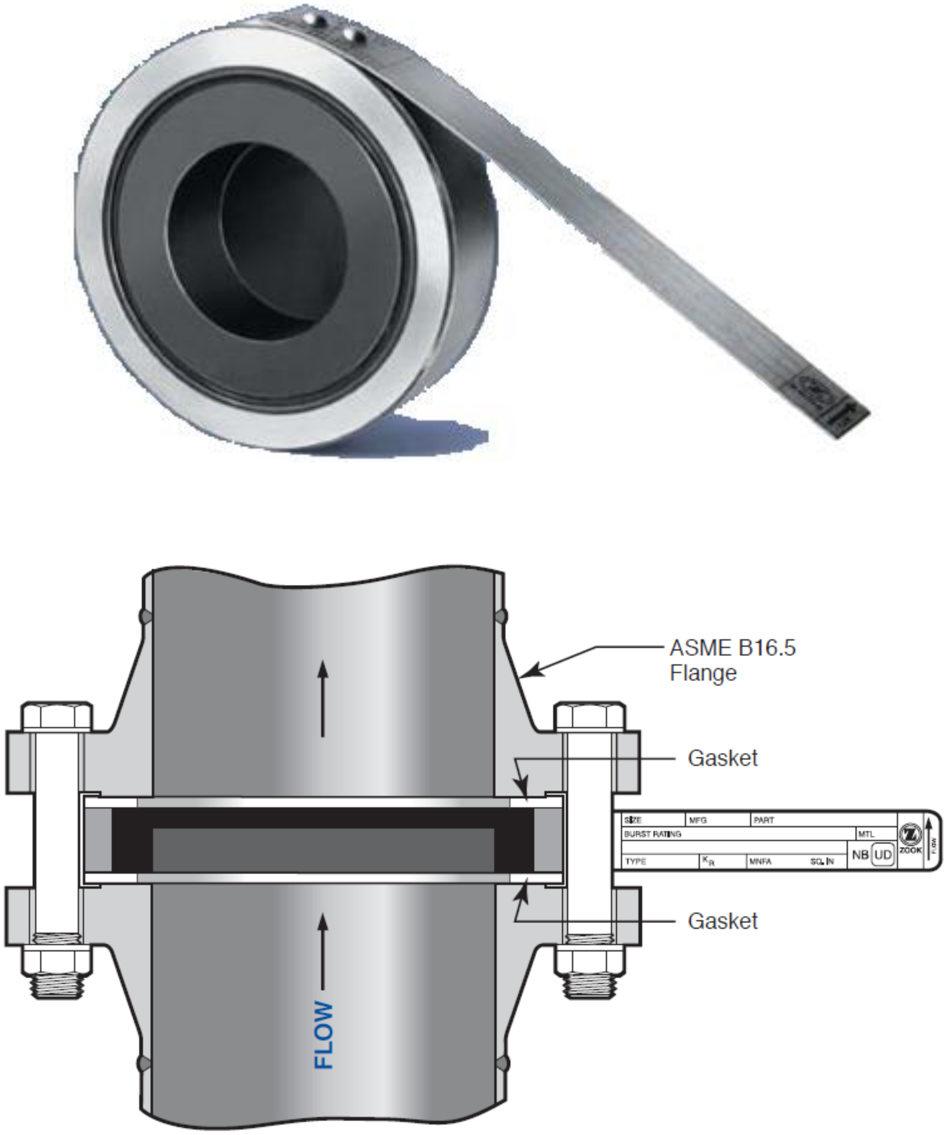


Figure 8. The rupture disk and assembly.

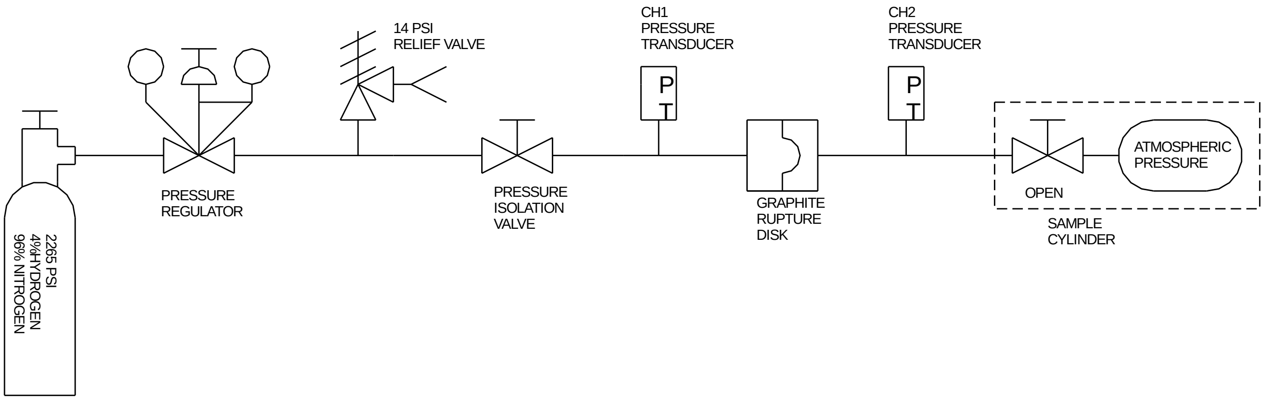


Figure 9. Test System Set-up for Analysis of the Rupture Disc.

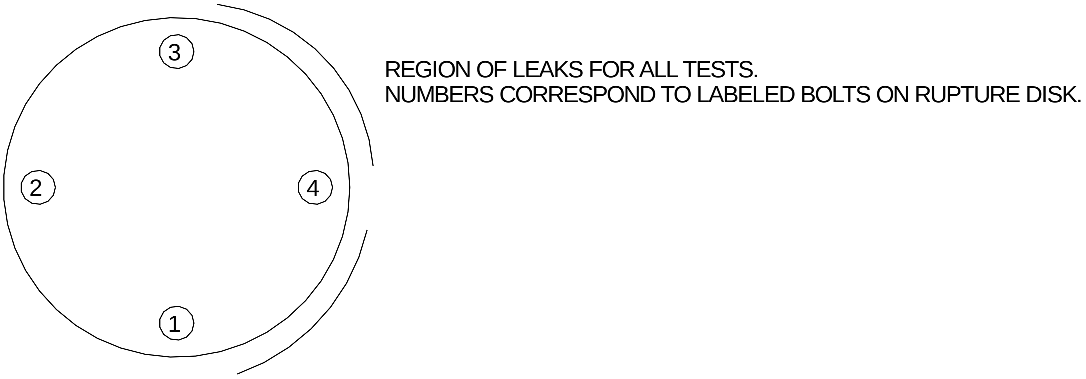


Figure 10. Labeled bolts on the rupture disc.

Tables

Irradiated Sample	Energy (KeV)	EOB DPM (% error)	Measured mCi in sample	Total mCi	Average mCi (STD)
Sample 2 – 2	181	272500000(4.7)	0.123	76.5	68(8)
Sample 2 – 5	181	220300000(4.6)	0.990	61.5	
Sample 2 – 6	181	232700000(5.1)	0.104	64.7	
Sample 2 – 2	740	207800000(3.0)	0.940	58.4	58(1)
Sample 2 – 5	740	200700000(3.1)	0.900	56.0	
Sample 2 – 6	740	208600000(3.3)	0.940	58.0	

Table 1. ^{99}Mo production values (181 and 740 KeV transitions) in irradiated LEU sulfate solution (DPM = disintegrations per minute). Data corrected to EOB (61.9167). The three samples were composed of a known mass of a sample (nominally 482 µL, with an accurate volume determined by density) of the 300.7 mL irradiated LEU sulfate solution, and 4500 µL of 0.1 mol L^{-1} H_2SO_4.

Radioisotope	Irradiated Sample	Sample volume measure/total (mL)	EOB DPM (% error)	Measured µCi in sample	Total mCi	Average(STD)
^{103}Ru	Sample 2 – 4	0.482/300.7	11560000(0.6)	5.21	3.25	3.26(1)
	Sample 2 – 5	0.485/300.7	11560000(0.6)	5.21	3.25	
	Sample 2 – 6	0.487/300.7	11660000(0.6)	5.25	3.27	
^{140}Ba	Sample 2 – 4	0.482/300.7	65440000(0.6)	29.5	18.38	18.32(5)
	Sample 2 – 5	0.485/300.7	65080000(0.6)	29.3	18.28	
	Sample 2 – 6	0.487/300.7	65110000(0.6)	29.3	18.29	
^{141}Ce	Sample 2 – 4	0.482/300.7	27450000(0.5)	12.4	7.71	7.73(4)
	Sample 2 – 5	0.485/300.7	27420000(0.5)	12.4	7.70	
	Sample 2 – 6	0.487/300.7	27690000(0.5)	12.5	7.78	

Table 2. ^{103}Ru (497 keV), ^{140}Ba (537 keV) and ^{141}Ce (145 keV) production values in irradiated LEU sulfate solution (DPM = disintegrations per minute). Data corrected to EOB (61.9167). The three samples were composed of a known mass of a sample (nominally 482 µL, with an accurate volume determined by density) of the 300.7 mL irradiated LEU sulfate solution, and 4500 µL of 0.1 mol L^{-1} H_2SO_4.

Radioisotope	Irradiated Sample	Sample volume measured/total (mL)	EOB DPM (% error)	Measured mCi in sample	Total mCi	Average(STD)
^{95}Zr	Sample 2 – 4	0.482/300.7	11030000(0.7)	0.00497	3.10	2.94(1)
	Sample 2 – 5	0.485/300.7	10120000(0.8)	0.00456	2.84	
	Sample 2 – 6	0.487/300.7	10300000(0.8)	0.00464	2.89	
^{147}Nd	Sample 2 – 4	0.482/300.7	27180000(0.6)	0.0122	7.63	7.65(12)
	Sample 2 – 5	0.485/300.7	26850000(0.6)	0.0121	7.54	
	Sample 2 – 6	0.487/300.7	27730000(0.6)	0.0125	7.79	
^{239}Np	Sample 2 – 4	0.482/300.7	418000000(3.1)	0.188	117	121(3)
	Sample 2 – 5	0.485/300.7	432900000(3.2)	0.195	122	
	Sample 2 – 6	0.487/300.7	442300000(3.1)	0.199	124	

Table 3. ^{95}Zr (757 keV), ^{147}Nd (91 keV) and ^{239}Np (278 keV) production values in irradiated LEU sulfate solution (DPM = disintegrations per minute). Data corrected to EOB (61.9167). The three samples were composed of a known mass of a sample (nominally 482 µL, with an accurate volume determined by density) of the 300.7 mL irradiated LEU sulfate solution, and 4500 µL of 0.1 mol L^{-1} H_2SO_4.

Radioisotope	Irradiated Sample	Sample volume measured/total (mL)	EOB DPM (% error)	Measured µCi in sample	Total mCi	Average(STD)
^{131}I	Sample 2 – 4	0.482/300.7	26150000(0.6)	11.78	7.34	7.39(4)
	Sample 2 – 5	0.485/300.7	26320000(0.6)	11.86	7.39	
	Sample 2 – 6	0.487/300.7	26440000(0.6)	11.91	7.43	

Table 4. ^{131}I (365 keV) production values in irradiated LEU sulfate solution (DPM = disintegrations per minute). Data corrected to EOB (61.9167). The three samples were composed of a known mass of a sample (nominally 482 µL, with an accurate volume determined by density) of the 300.7 mL irradiated LEU sulfate solution, and 4500 µL of 0.1 mol L^{-1} H_2SO_4. At EOB the % contribution to ^{131}I activity from $^{131m/131}$Te ingrowth would be <5%.

	Run 1, g (mL)	Run 2, g (mL)
0.1 mol L^{-1} H_2SO_4 conditioning wash	57 (57)	49 (49)
Irradiated LEU feed solution	171.4 (144.5)	164.8 (139.0)
1.0 mol L^{-1} H_2SO_4 wash	10.6 (10.0)	14.2 (13.4)
H_2O wash	23.0 (23.0)	24.6 (24.6)
0.1 mol L^{-1} NaOH	45 (45)	43 (43)

Table 5. Column separation experiments – input solution mass (g) and volume (mL).

	Run 1, g (mL)	Run 2, g (mL)
0.1 mol L^{-1} H_2SO_4 wash	56.4	49.8
Irradiated LEU Fraction	168.6 (142.7)	163.0 (138.5)
1.0 mol L^{-1} H_2SO_4 and H_2O wash	32.0	38.9
0.1 mol L^{-1} NaOH	50.0 (50.0)	39.2 (39.2)

Table 6. Column separation experiments – collected solution fraction masses (g) and, where densities were measured, volumes (mL).

	Samples	Sample volume measured/total (mL)	EOB sample DPM (% error)	EOB mCi	Column end mCi	% activity balance
Separated LEU run 1	Irrad. LEU pre-separation				5.01	
	Sample 3	0.4862/141.68	63970000(0.7)	8.40	4.88	97
	Sample 6 (1-2)	0.4871/141.68	64230000(0.7)	8.42	4.89	98
	Sample 7 (1-3)	0.4853/141.68	63560000(0.7)	8.36	4.86	97
Separated LEU run 2	Irrad. LEU pre-separation				4.59	
	Sample 10 (2-1)	0.4894/137.11	63550000(0.7)	8.02	4.41	96
	Sample 2 – 2	0.4880/137.11	63100000(0.7)	7.99	4.39	96
	Sample 2 – 3	0.4889/137.11	64140000(0.7)	8.10	4.45	97

Table 7. ^{140}Ba (537 keV) activity values in the two separated LEU sulfate fractions collected after the titania column separations at the CMR (DPM = disintegrations per minute). Data corrected to both EOB (61.9167) and column end time (Separated LEU run 1 and 2 = 70.9271 and 71.8507, respectively). All the reported mCi measurements are calculated to the sample volume of irradiated LEU used for the specific column separation experiment (141.68 mL for the first run and 137.11 mL for the second run). The three samples were composed of a known mass of a sample (nominally 482 µL, with an accurate volume determined by density) and 4500 µL of 0.1 mol L^{-1} H_2SO_4.

	Samples	Sample volume measured/total (mL)	EOB sample DPM (% error)	EOB mCi	Column end mCi	% activity balance
Separated LEU run 1	Irrad. LEU pre-separation				3.01	
	Sample 3	0.4862/141.68	27160000(0.5)	3.57	2.94	98
	Sample 6 (1-2)	0.4871/141.68	27430000(0.5)	3.59	2.97	99
	Sample 7 (1-3)	0.4853/141.68	27030000(0.5)	3.55	2.93	97
Separated LEU run 2	Irrad. LEU pre-separation				2.91	
	Sample 10 (2-1)	0.4894/137.11	27260000(0.5)	3.44	2.78	98
	Sample 2 – 2	0.4880/137.11	27910000(0.5)	3.53	2.86	100
	Sample 2 – 3	0.4889/137.11	27150000(0.5)	3.43	2.78	97

Table 8. ^{141}Ce (145 keV) activity values in the two separated LEU sulfate fractions collected after the titania column separations at the CMR (DPM = disintegrations per minute). Data corrected to both EOB (61.9167) and column end time (Separated LEU run 1 and 2 = 70.9271 and 71.8507, respectively). All the reported mCi measurements are calculated to the sample volume of irradiated LEU used for the specific column separation experiment (141.68 mL for the first run and 137.11 mL for the second run). The three samples were composed of a known mass of a sample (nominally 482 µL, with an accurate volume determined by density) and 4500 µL of 0.1 mol L^{-1} H_2SO_4.

	Samples	Sample volume measured/total (mL)	EOB sample DPM (% error)	EOB mCi	Column end mCi	% activity balance
Separated LEU run 1	Irrad. LEU pre-separation				2.04	
	Sample 3	0.4862/141.68	26540000(0.7)	3.48	1.97	97
	Sample 6 (1-2)	0.4871/141.68	26870000(0.7)	3.53	2.00	98
	Sample 7 (1-3)	0.4853/141.68	26080000(0.7)	3.42	1.94	95
Separated LEU run 2	Irrad. LEU pre-separation				1.86	
	Sample 10 (2-1)	0.4894/137.11	26400000(0.7)	3.33	1.78	96
	Sample 2 – 2	0.4880/137.11	26190000(0.7)	3.31	1.77	95
	Sample 2 – 3	0.4889/137.11	26420000(0.7)	3.33	1.78	96

Table 9. ^{147}Nd (91 keV) activity values in the two separated LEU sulfate fractions collected after the titania column separations at the CMR (DPM = disintegrations per minute). Data corrected to both EOB (61.9167) and column end time (Separated LEU run 1 and 2 = 70.9271 and 71.8507, respectively). All the reported mCi measurements are calculated to the sample volume of irradiated LEU used for the specific column separation experiment (141.68 mL for the first run and 137.11 mL for the second run). The three samples were composed of a known mass of a sample (nominally 482 µL, with an accurate volume determined by density) and 4500 µL of 0.1 mol L^{-1} H_2SO_4.

	Samples	Sample volume measured/total (mL)	EOB sample DPM (% error)	EOB mCi	Column end mCi	% activity balance
Separated LEU run 1	Irrad. LEU pre-separation				4.02	
	Sample 3	0.4862/141.68	387800000(4.0)	50.9	3.59	89
	Sample 6 (1-2)	0.4871/141.68	415400000(3.7)	54.5	3.84	96
	Sample 7 (1-3)	0.4853/141.68	402700000(3.9)	52.9	3.73	93
Separated LEU run 2	Irrad. LEU pre-separation				2.96	
	Sample 10 (2-1)	0.4894/137.11	384700000(4.1)	48.5	2.61	88
	Sample 2 – 2	0.4880/137.11	355200000(4.1)	44.8	2.41	81
	Sample 2 – 3	0.4889/137.11	420800000(4.0)	53.1	2.85	96

Table 10. ^{239}Np (278 keV) activity values in the two separated LEU sulfate fractions collected after the titania column separations at the CMR (DPM = disintegrations per minute). Data corrected to both EOB (61.9167) and column end time (Separated LEU run 1 and 2 = 70.9271 and 71.8507, respectively). All the reported mCi measurements are calculated to the sample volume of irradiated LEU used for the specific column separation experiment (141.68 mL for the first run and 137.11 mL for the second run). The three samples were composed of a known mass of a sample (nominally 482 µL, with an accurate volume determined by density) and 4500 µL of 0.1 mol L^{-1} H_2SO_4.

	Samples	Sample volume measured/total (mL)	EOB sample DPM (% error)	EOB mCi	Column end mCi	% activity balance
Separated LEU run 1	Irrad. LEU pre-separation				1.31	
	Sample 3	0.4862/141.68	10850000(0.7)	1.42	1.21	92
	Sample 6 (1-2)	0.4871/141.68	10820000(0.7)	1.42	1.21	92
	Sample 7 (1-3)	0.4853/141.68	10810000(0.7)	1.42	1.21	92
Separated LEU run 2	Irrad. LEU pre-separation				1.25	
	Sample 10 (2-1)	0.4894/137.11	10690000(0.7)	1.35	1.13	90
	Sample 2 – 2	0.4880/137.11	10600000(0.7)	1.34	1.13	90
	Sample 2 – 3	0.4889/137.11	10680000(0.7)	1.35	1.13	90

Table 11. ^{103}Ru (497 keV) activity values in the two separated LEU sulfate fractions collected after the titania column separations at the CMR (DPM = disintegrations per minute). Data corrected to both EOB (61.9167) and column end time (Separated LEU run 1 and 2 = 70.9271 and 71.8507, respectively). All the reported mCi measurements are calculated to the sample volume of irradiated LEU used for the specific column separation experiment (141.68 mL for the first run and 137.11 mL for the second run). The three samples were composed of a known mass of a sample (nominally 482 µL, with an accurate volume determined by density) and 4500 µL of 0.1 mol L^{-1} H_2SO_4.

	Samples	Sample volume measured/total (mL)	EOB sample DPM (% error)	EOB mCi	Column end mCi	% activity balance
Base fraction run 1	Irrad. LEU pre-separation				3.30	
	Sample 5 Mo/NaOH	0.993/49.8	1196000000(1.0)	27.0	2.78	84
	Sample 5 Mo/NaOH	0.989/49.8	1223000064(1.0)	27.7	2.86	87
	Sample 5 Mo/NaOH	0.995/49.8	1206000000(1.0)	27.2	2.80	85
Base fraction run 2	Irrad. LEU pre-separation				2.53	
	9 sample 1 – 5	0.990/39.1	1411000064(0.9)	25.0	2.04	81
	9 sample 1 – 5	0.993/39.1	1384000000(0.9)	24.6	2.01	79
	9 sample 1 – 5	0.993/39.1	1411000064(0.9)	25.0	2.04	81

Table 12. ^{99}Mo (181 keV) activity values in the two NaOH 0.1 mol L^{-1} base fractions collected after the titania column separations at the CMR (DPM = disintegrations per minute). Data corrected to both EOB (61.9167) and column end time (Separated LEU run 1 and 2 = 70.9271 and 71.8507, respectively). All the reported mCi measurements are calculated to the sample volume of base used for the specific column separation experiment (49.8 mL for the first run and 39.1 mL for the second run). The three samples were composed of a known mass of a sample (nominally ca. 1000 µL, with an accurate volume determined by density) and 4000 µL of 0.1 mol L^{-1} NaOH.

	Samples	Sample volume measured/total (mL)	EOB sample DPM (% error)	EOB mCi	Column end mCi	% activity balance
Base fraction run 1	Irrad. LEU pre-separation				2.81	
	Sample 5 Mo/NaOH	0.993/49.8	1151000064(1.3)	26.0	2.68	95
	Sample 5 Mo/NaOH	0.989/49.8	1143000064(1.3)	25.9	2.67	95
	Sample 5 Mo/NaOH	0.995/49.8	1148999936(1.0)	25.9	2.67	95
Base fraction run 2	Irrad. LEU pre-separation				2.16	
	9 sample 1 – 5	0.990/39.1	1342000000(1.2)	23.8	1.94	91
	9 sample 1 – 5	0.993/39.1	1332000000(1.3)	23.7	1.94	90
	9 sample 1 – 5	0.993/39.1	1334000000(1.0)	23.7	1.93	90

Table 13. ^{99}Mo (740 keV) activity values in the two NaOH 0.1 mol L^{-1} base fractions collected after the titania column separations at the CMR (DPM = disintegrations per minute). Data corrected to both EOB (61.9167) and column end time (Separated LEU run 1 and 2 = 70.9271 and 71.8507, respectively). All the reported mCi measurements are calculated to the sample volume of base used for the specific column separation experiment (49.8 mL for the first run and 39.1 mL for the second run). The three samples were composed of a known mass of a sample (nominally ca. 1000 µL, with an accurate volume determined by density) and 4000 µL of 0.1 mol L^{-1} NaOH.

	Samples	**Sample volume measured/total (mL)**	**EOB sample DPM (% error)**	**EOB mCi**	**Column end mCi**	**% activity balance**
Separated LEU run 1	Irrad. LEU pre-separation				1.60	
	Sample 3	0.4862/141.68	12940000(0.9)	1.70	0.780	49
	Sample 6 (1-2)	0.4871/141.68	12680000(0.9)	1.66	0.763	48
	Sample 7 (1-3)	0.4853/141.68	12750000(0.9)	1.68	0.769	48
Separated LEU run 2	Irrad. LEU pre-separation				1.43	
	Sample 10 (2-1)	0.4894/137.11	12030000(1.0)	1.52	0.643	45
	Sample 2 – 2	0.4880/137.11	11930000(1.0)	1.51	0.640	45
	Sample 2 – 3	0.4889/137.11	11750000(1.0)	1.48	0.629	44

Table 14. ^{131}I (365 keV) activity values in the two separated LEU sulfate fractions collected after the titania column separations at the CMR (DPM = disintegrations per minute). Data corrected to both EOB (61.9167) and column end time (Separated LEU run 1 and 2 = 70.9271 and 71.8507, respectively). All the reported mCi measurements are calculated to the sample volume of irradiated LEU used for the specific column separation experiment (141.68 mL for the first run and 137.11 mL for the second run). The three samples were composed of a known mass of a sample (nominally 482 µL, with an accurate volume determined by density) and 4500 µL of 0.1 mol L^{-1} H_2SO_4.

	Samples	Sample volume measured/total (mL)	EOB sample DPM (% error)	EOB mCi	Column end mCi	% activity balance
Base fraction run 1	Irrad. LEU pre-separation				1.61	
	Sample 5 Mo/NaOH	0.993/49.8	80400000(0.7)	1.82	0.834	52
	Sample 5 Mo/NaOH	0.989/49.8	80010000(0.6)	1.81	0.833	52
	Sample 5 Mo/NaOH	0.995/49.8	81160000(0.6)	1.82	0.840	53
Base fraction run 2	Irrad. LEU pre-separation				1.49	
	9 sample 1 – 5	0.990/39.1	58710000(0.7)	1.04	0.461	31
	9 sample 1 – 5	0.993/39.1	58530000(0.7)	1.04	0.461	31
	9 sample 1 – 5	0.993/39.1	58250000(0.7)	1.03	0.457	31

Table 15. ^{131}I (365 keV) activity values in the two NaOH 0.1 mol L^{-1} base fractions collected after the titania column separations at the CMR (DPM = disintegrations per minute). Data corrected to both EOB (61.9167) and column end time (Separated LEU run 1 and 2 = 70.9271 and 71.8507, respectively). All the reported mCi measurements are calculated to the sample volume of base used for the specific column separation experiment (49.8 mL for the first run and 39.1 mL for the second run). The three samples were composed of a known mass of a sample (nominally ca. 1000 µL, with an accurate volume determined by density) and 4000 µL of 0.1 mol L^{-1} NaOH.

Test 1 (5 ft-lb torque)

TIME (min.)	CH1 (Torr)	CH2 (Torr)	LEAK RATE (Torr/min)
16:15	11.387	11.380	-
16:34	14.56	11.379	- (pressure increase)
16:51	12.817	11.379	1.743
18:20	11.404	11.379	1.413

Test 2 (5-ft-lb torque)

TIME (min.)	CH1 (Torr)	CH2 (Torr)	LEAK RATE (Torr/min)
0	14.6	11.405	-
1:00	14.424	11.405	.176
2:00	14.278	11.402	.146
3:00	14.132	11.403	.133
4:00	13.999	11.402	.127

Test 3 (5 ft-lb torque)

TIME (min.)	CH1 (Torr)	CH2 (Torr)	LEAK RATE (Torr/min)
11:00	13.240	11.401	.105 (unsubstantiated)
15:00	12.920	11.401	.080
17:00	12.773	11.398	.074
19:00	12.650	11.398	.062
21:00	12.535	11.398	.058

Table 16. First set of leak test data for the rupture discs.

Test 4 (17 ft-lb torque)

TIME (min.)	CH1 (Torr)	CH2 (Torr)	LEAK RATE (Torr/min)
0	14.425	11.425	-
1	14.200	11.423	.225
2	14.034	11.425	.166
3	13.871	11.423	.163
4	13.727	11.423	.144
5	13.584	11.423	.143

Test 5 (17 ft-lb torque)

TIME (min.)	CH1 (Torr)	CH2 (Torr)	LEAK RATE Torr/min
0	14.527	11.421	-
1	14.376	11.422	.151
2	14.222	11.422	.154
3	14.080	11.422	.142
4	13.947	11.421	.133
5	13.814	11.421	.133

Test 6 (17 ft-lb torque)

TIME (min.)	CH1 (Torr)	CH2 (Torr)	LEAK RATE Torr/min
0	14.454	11.421	-
1	14.293	11.420	.161
2	14.148	11.420	.145
3	14.007	11.420	.141
4	13.876	11.420	.131
5	13.752	11.419	.124

Table 17. Second set of leak test data for the rupture discs.