

# Mo-99 2014 TOPICAL MEETING ON MOLYBDENUM-99 TECHNOLOGICAL DEVELOPMENT

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## Overview: Argonne Assistance in Developing SHINE Production of Mo-99

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### ABSTRACT

Argonne National Laboratory is providing technical assistance to SHINE Medical Technologies in their efforts to become a domestic Mo-99 producer. Significant progress has been made in successfully producing, separating, and purifying 1-2 Ci of Mo-99 using Argonne's linac, 5 L of low enriched uranyl sulfate solution, and the mini-SHINE experimental setup. Turbidity, pH, and conductivity are continuously monitored during irradiation, and gas generation rates and fission product speciation have been measured during each mini-SHINE run. Important decisions related to centrifugal contactor design and target solution cleanup have been made, and as a result, considerable advancements in the areas of plant design and waste optimization have occurred. Argonne has reviewed SHINE process flow diagrams for Mo recovery, purification, and periodic target-solution cleanup and optimized potential waste streams to minimize costs and meet specifications for radioactive-waste disposal. Additionally, modeling and experimentation associated with bubble formation and precipitation have been examined. This paper provides an overview of Argonne progress in developing SHINE's process for Mo-99 production.

### Introduction

Argonne National Laboratory is assisting SHINE Medical Technologies in their efforts to become a domestic Mo-99 producer. The mini-SHINE experimental setup is a complex system, which includes a gas collection and analysis system, target solution monitoring glovebox where pH, conductivity, and turbidity are continuously being monitored during irradiation, Mo-recovery glovebox, shielded cell that houses the target solution, and a separate hot cell used for the concentration column, alkaline-to-acidic conversion, and LEU-Modified Cintichem purification processes. Samples are collected during irradiation, the initial Mo-recovery column, and all secondary purification steps performed in a separate hot cell. Samples collected during all processing steps are retrieved and gamma counted to determine Mo-recovery yields, fission production partitioning, and purity specifications.

SHINE plans to clean-up their target solution every 4 or more irradiation cycles using the UREX process [1]. The first step involves conversion of uranyl sulfate to uranyl nitrate using a combination of strontium and barium nitrate to form uranyl nitrate and precipitate strontium and barium sulfate. The filtrate and nitric acid washes will enter the UREX process to remove fission and activation products that have built up during the course of 4 or more irradiation cycles. The size of the centrifugal contactors that would be best suited for SHINE's process volume of 240-300-L is the "V-3.5," which has a rotor diameter of 3.5 inches—9 cm. Given the reduced hold-up, cold-feed requirements and waste volumes, the "V-3.5" would appear to be a nearly optimal scale for this volume [2].

Target solution cleanup generates the majority of SHINE's waste, and Mo-99 separation, recovery, and purification steps generate another portion of the waste. Equipment and components from the gas-collection system will also contribute to SHINE's waste streams, but the replacement frequency for these components is currently unknown. A study is currently underway to review the SHINE process flowsheets, add assumptions about partitioning of components based on the chemistry developed at Argonne, and develop volumes and classifications of all waste streams based on an entire mass balance of the facility.

SHINE Medical Technologies needs data to assure themselves and the Nuclear Regulatory Commission (NRC) that precipitate formation will not occur in their 140 g-U/L uranyl sulfate target solution. Potential sources of precipitation include uranyl peroxide, fission products, and corrosion products. Formation of uranyl-peroxide can be mitigated by adding 1 ppm  $\text{FeSO}_4$  prior to irradiation to catalyze the destruction of peroxide before it can build up enough to cause precipitation. This has been verified with irradiation tests performed at the Van de Graaff and phase-1 micro-SHINE irradiation tests performed using the linac [3,4]. Chemical modeling of the SHINE target solution showed that certain phases could form, but their precipitation kinetics are unknown [5]. As a result, the potential for precipitation of  $\text{ZrO}_2$ ,  $\text{SnO}_2$ ,  $\text{BaSO}_4$ ,  $\text{CoWO}_4$ , and  $\text{RuO}_2$  in a uranyl sulfate solution at room temperature and  $60^\circ\text{C}$  has been examined experimentally [6]. Small-scale corrosion tests were performed at Argonne using 304SS and 316SS disks in contact with uranyl sulfate at room temperature,  $60^\circ\text{C}$ , and  $80^\circ\text{C}$  [7]. No precipitation of any potential corrosion products (Fe, Co, Cr, and Ni) was observed during these tests which had incubation times of 6 hours to 30 days. ICP-MS results indicate that corrosion rates are high at first, but once a passivation layer forms, corrosion rates become negligible.

The electron beam of the linac will be used to irradiate a solution volume (sector of the target solution vessel) to study radiolytic-bubble formation and the thermal hydraulics of the system. Experimental results obtained will be compared with simulations to fine tune the computer models developed by SHINE and Los Alamos National Laboratory. Because the solution will undergo radiolytic decomposition in this experiment, bubble formation and dynamics can be studied along with the effects of convection and temperature on bubble behavior. These experiments will serve as a verification/validation tool for the thermal-hydraulic models. While data on radiolytic gas formation will be collected in mini-SHINE experiments, bubble formation dynamics data will be limited due to the complexity of the optical setup for the extremely high radiation fields in the fissioning solution. Utilization of the direct electron beam for irradiation allows homogenous heating of a large solution volume and simplifies observation of the bubble dynamics simultaneously with thermal-hydraulic data collection.

A significant amount of work being done to support the SHINE project utilizes Argonne's linac. As progress is made especially with the mini-SHINE experiments at the linac, new modifications

and/or additional tests are required. For example, significant color changes were observed after the sodium bisulfate solution contacted the stainless steel tubing in the mini-SHINE setup. As a result, a series of corrosion tests were performed to ensure that corrosion would not continue to increase after several days of contact. Mo-99 for shipment has not been produced yet, but a significant amount of progress has been made and a lot of lessons have been learned that will be extremely valuable to SHINE during their building and startup phases.

## **Experimental**

### *Mini-SHINE Irradiations*

To date, water and sodium bisulfate solutions spiked with Mo-99 and stable Mo have been irradiated using Argonne's linac. Approximately 5 L of each solution were irradiated in a 304SS target solution vessel in a shielded cell. During each irradiation, pH, conductivity, and turbidity were constantly monitored. In addition, gases generated during radiolysis were analyzed using a GC-MS (gas chromatography-mass spectrometer), RGA (residual gas analyzer), and H<sub>2</sub> sensor. Sodium bisulfate solutions containing a Mo-99 spike were passed through a titania column after irradiation, and the Mo-product was transferred to a second hot cell for further processing. All glovebox operations are performed remotely using LabView software. Samples are retrieved manually at least 8 hours after irradiation. The Mo-product was acidified and passed through a second concentration column. The Mo-product from the concentration column was evaporated twice and acidified before entering the LEU-Modified Cintichem purification process.

### *Preparation of Mo-99 Spike Solution.*

Mo-99 is removed from a Tc-99m generator by placing a serum vial containing 1 M NH<sub>4</sub>OH on the needle labeled "Saline Charge". Then an evacuated serum vial is placed on the needle labeled "Receiver". The Mo-99 spiked solution is prepared by bringing the solution to dryness on a hot plate, and re-dissolving it in 0.1 M H<sub>2</sub>SO<sub>4</sub> or H<sub>2</sub>O.

### *Counting of Mo-99*

The amount of activity in the aqueous samples is determined using a germanium detector. Mo-99 is quantified by measurement of its 739 keV  $\gamma$ -ray. The activity of Mo-99 in each sample is corrected for decay.

## **Results and Discussion**

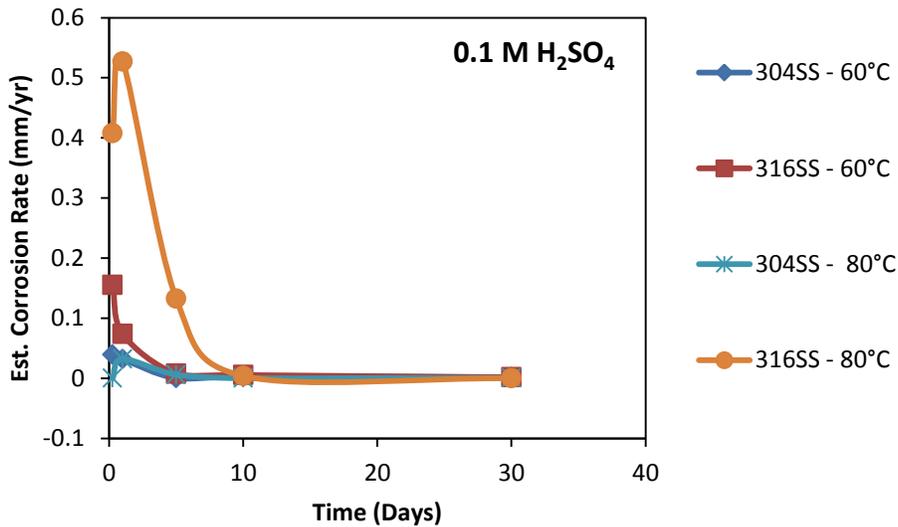
### *Mini-SHINE Experiments*

A series of 5-L water irradiations and phase-1 micro-SHINE irradiations were performed in FY13 [4]. In FY14, several runs of the Mo-recovery system and concentration and purification procedures were performed using a sodium-bisulfate solution with and without irradiation. Due to the fact that the sodium bisulfate solution turned blue after contacting the stainless steel tubing in the mini-SHINE setup, a series of corrosion tests were performed.

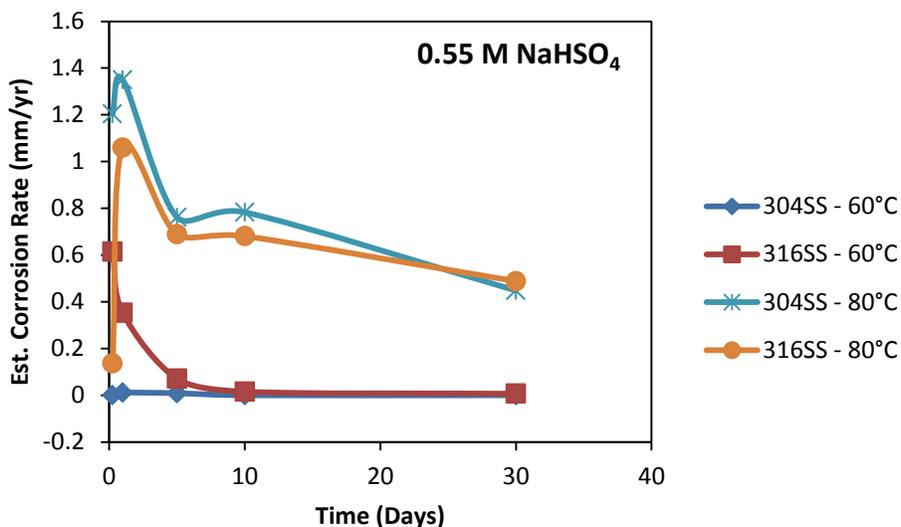
After passing a 0.55 M NaHSO<sub>4</sub>/0.1 M H<sub>2</sub>SO<sub>4</sub> (pH 1) solution through the target solution vessel and stainless-steel tubing, valves, and pump as part of the mini-SHINE experimental setup at the linac, the solution turned blue. The blue color intensified when the system temperature reached 80°C. The tubing and mini-SHINE target-solution vessel are made of 304SS, and all other components are made of 316SS. As a result, corrosion tests were performed using 304SS and 316SS in contact with H<sub>2</sub>O, 0.1 M H<sub>2</sub>SO<sub>4</sub>, 0.55 M NaHSO<sub>4</sub>, and 140 g-U/L UO<sub>2</sub>SO<sub>4</sub> for 6 hours,

24 hours, 5 days, 10 days, and 30 days at 60°C and 80°C [6]. Corrosion-rate data were required to ensure the sodium bisulfate, sulfuric acid (pH = 1), and uranyl sulfate (140 g-U/L) solutions would not corrode the components of the mini-SHINE setup to any great extent.

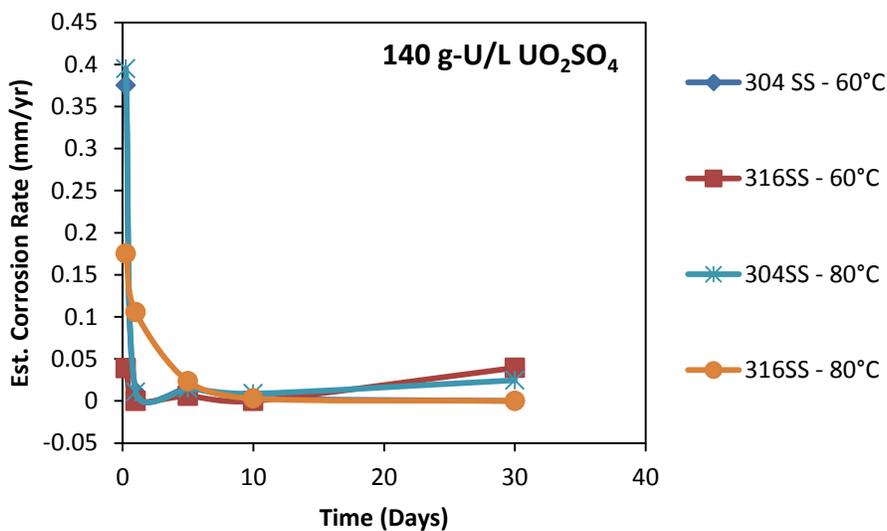
In general, the discs in contact with 0.1 M H<sub>2</sub>SO<sub>4</sub> (Figure 1) have lower estimated corrosion rates than the discs that were contacted with 0.55 M NaHSO<sub>4</sub> (Figure 2) and 140 g-U/L UO<sub>2</sub>SO<sub>4</sub> (Figure 3). The 316SS samples show the highest initial corrosion rates after 6 and 24 hours of contact with 0.1 M H<sub>2</sub>SO<sub>4</sub> (Figure 1); however, after 10 days, the estimated corrosion rates decrease significantly. Estimated corrosion rates for all samples in contact with 0.1 M H<sub>2</sub>SO<sub>4</sub> become negligible after 10 days. Sodium bisulfate is the most corrosive solution, as it has the highest concentration of free sulfate. Titanium metal is most affected by 0.55 M NaHSO<sub>4</sub> (Figure 2) where corrosion rates at 80°C do not fall below 3 mm/yr. Both 304SS and 316SS show corrosion rates at 80°C of 1-1.2 mm/yr after contact with 0.55 M NaHSO<sub>4</sub>, but those rates decrease to 0.5 mm/yr after 30 days. Uranyl sulfate is slightly more corrosive than 0.1 M H<sub>2</sub>SO<sub>4</sub>, but corrosion rates are below 0.05 mm/yr for all discs after 30 days (Figure 3).



**Figure 1. Estimated corrosion rates in units of mm/yr for discs in contact with 0.1 M H<sub>2</sub>SO<sub>4</sub> at 60°C and 80°C.**



**Figure 2. Estimated corrosion rates in units of mm/yr for discs in contact with 0.55 M NaHSO<sub>4</sub> at 60°C and 80°C.**



**Figure 3. Estimated corrosion rates in units of mm/yr for discs in contact with 140 g-U/L UO<sub>2</sub>SO<sub>4</sub> at 60°C and 80°C.**

Stable Mo (3  $\mu$ M) was added as sodium molybdate and 2 – 10 mCi Mo-99 was added for each run. Table 1 shows the results from the various mini-SHINE runs using a 0.55 M NaHSO<sub>4</sub>, pH 1 solution. The experiments performed to date show lower than expected Mo-recovery yields after the first titania column. Typically, we would expect >90% of the Mo recovered after the first column, but the missing ~20% (except for the run performed on 03/26/14) Mo was not found in the effluent, in the washes, or on the column. The low initial Mo yield is currently being investigated and may be due to plating out or precipitation in the coils inside the heater blocks or stainless steel tubing lines that are exposed to acid and base solutions. Mo-recovery after the

concentration column is in the expected range except for the first run performed after the solution was irradiated; however, the second run performed after a longer irradiation (04/15/14) showed recoveries in the expected ranges. Most of the recoveries after the LEU-Modified Cintichem process were lower than expected due to the Mo-product bottle breaking and insufficient removal of the Mo-product after evaporation. Modifications to the setup have been made to help prevent the Mo-product bottle from breaking, and a new flask is currently being used to help washing after evaporation. A long irradiation with sodium bisulfate is planned for the week of June 9<sup>th</sup>, and after successful completion of that experiment, uranyl sulfate will be irradiated to produce 1-2 Ci Mo-99 for shipment to a Tc-99m generator manufacturer for testing.

Table 1. %Mo recovered during each step for the mini-SHINE runs with NaHSO<sub>4</sub> and Mo-99.

Date of Experiment	Irradiation	Irradiation Time (min)	1st Column	2nd Column	Cintichem
2/3/2014	no	NA	80%	88%	84%
3/10/2014	no	NA	88%	96%	62%
3/26/2014	yes	80	48%	82%	64%*
4/15/2014	yes	300	80%	95%	70%**

\* % Mo recovered after evaporation – Mo-product bottle broke

\*\* % Mo recovered after evaporation and Cintichem

Gas data are collected during each irradiation as well using a GC-MS, RGA, and H<sub>2</sub> sensor. The H<sub>2</sub> sensor and H<sub>2</sub>/O<sub>2</sub> catalytic recombiner failed during the irradiation performed on 04/15/14 because both got wet from condensation in the system. The TCD (thermal conductivity detector – part of the GC) results are shown in Figure 4. The beam was turned off because the RGA failed early around 12:00, and the H<sub>2</sub> sensor failed around 15:00. The GC-TCD was the only way to measure H<sub>2</sub>, and it has a delay time of several minutes, so it was decided to terminate the run at 15:02. Modifications have been made to the system to prevent failure of the catalyst, H<sub>2</sub> sensor, and RGA output. A new condenser has been designed and installed to minimize liquid in the system, and the catalytic recombiner has been wrapped with heat tape. Another issue that has been addressed is the performance of the RGA in high radiation area. More shielding has been installed, and in addition to the already existing digital output to the computer on the RGA, four analog outputs have been wired to separate data loggers. The analog outputs are expected to withstand the high radiation fields better than the digital outputs.

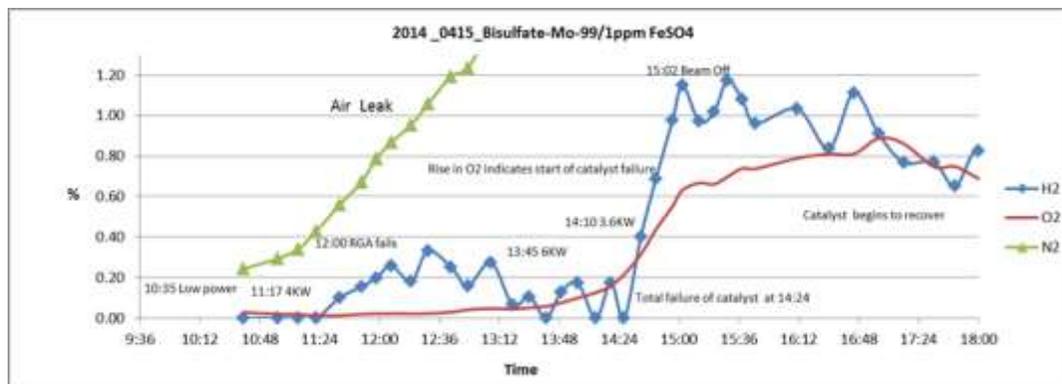


Figure 4. GC-TCD data for sodium bisulfate irradiation performed on 04/15/14.

### *Contactors Design and Waste*

Originally, Argonne and SHINE were focusing on two different contactor designs sold by CINC Industries (<http://www.cincind.com/>) for use in the UREX treatment of spent target solutions. The V-2 (2-inch rotor diameter) requires a substantially longer processing time, up to 19 hours. The V-5 (5-inch rotor diameter) reduces the processing time by a factor of nine, but the volumes of waste that are generated go up by a factor of ten or more. An intermediate contactor size, designated "V-3.5," based on the results of the earlier study focusing on the V-2 and V-5, additional information obtained from SHINE Medical Technologies, process flow diagrams for the Mo-99 production process, and discussions with CINC Industries has been generated [1]. The quantities of cold chemicals needed simply to start-up and shut down the V-5 are more than halved for the "V-3.5." On the other hand, the processing time required for the "V-3.5" is approximately double. The length of the of a 30-stage "V-3.5" contactor bank is less than of that for the V-5; additionally, there is some room for reducing the calculated lengths by about 20% based on the unit installed in the MCU facility at SRS. The smaller "V-3.5" would free up additional shielded-cell space for other operations such as solvent clean-up or equipment maintenance, as well as any unforeseen tasks.

Perhaps the most significant determinant in the initial evaluation was the processing time. For the feed batch volume of 260 L used in the first calculation, the V-2 requires 19 hours which was deemed too long to allow completion in a single day, while the V-5 requires approximately 3 hours, and the "V-3.5" requires 5.7 hours. Given the reduced hold-up, cold-feed requirements, and waste volumes, the "V-3.5" would appear to be a nearly optimal scale for the 260-L feed batch volume. It should be emphasized that the characteristics of the "V-3.5" are derived from the calculations made for the V-2 and V-5 where key design parameters were available for input to the ROTOR model to predict contactor operational characteristics. Because these data are not yet available for the "V-3.5," the predicted values are more uncertain, and processing time, hold-up, and footprint will be somewhat, perhaps significantly, different than shown here.

Work is currently ongoing to determine the best way to handle SHINE's waste. A final report discussing Argonne's results will be available in September 2014.

### *Precipitation: Models and Experimentation*

The following results are from modeling the chemistry of the target solution using the code called The Geochemist's Workbench®, Professional Release 8.0 (GWB) [5]. The saturation state of the solution is calculated with respect to a large data base of potential phases to identify solid phases that are thermodynamically stable (candidates for possible precipitation) under relevant conditions (temperature, pH, Eh, etc.). The aqueous speciation of the solution is also described, as this information can be used to predict the chemical evolution of the system (pH, Eh, saturation state) if conditions change (temperature, solution composition). The solution is predicted to be saturated with respect to several solid phases, although some of these are not expected to form due to slow precipitation kinetics. Some solids with unknown precipitation kinetics that have been investigated experimentally are ZrO<sub>2</sub>, SnO<sub>2</sub>, BaSO<sub>4</sub>, CoWO<sub>4</sub>, and RuO<sub>2</sub>. Zr, added as Zr(SO<sub>4</sub>)<sub>2</sub>·4H<sub>2</sub>O, did not precipitate in the presence of a 140 g-U/L uranyl sulfate solution, pH 1 at concentrations as high as 30 mM at room temperature and 60°C. Precipitation was not observed in solutions containing 30 mM Zr after being left for one month at room temperature and 60°C.

Sn, added as  $\text{SnCl}_4 \cdot 5\text{H}_2\text{O}$ , showed no signs of precipitation when the final Sn concentration was 0.01 mM, 0.1 mM, 1 mM, 3 mM, 10 mM, or 20 mM; however, when the final Sn concentration reached 30 mM in the presence of a 140 g-U/L uranyl sulfate solution, pH 1, instantaneous precipitation occurred at room temperature and 60°C. Solutions containing 20 mM Sn at room temperature and 60°C had precipitates after 2 weeks, but no precipitation occurred in solutions containing 0.01 – 10 mM Sn after 1 month.

Ba, added as  $\text{Ba}(\text{NO}_3)_2$ , was soluble in the presence of a 140 g-U/L uranyl sulfate solution, pH 1 at concentrations of 0.01 mM and 0.1 mM, but when the concentration reached 1 mM Ba at room temperature and 60°C, a precipitate formed immediately. After one month, precipitates did not form in solutions containing 0.01 mM and 0.1 mM Ba at room temperature and 60°C.

$\text{CoWO}_4$ , added as two separate metal salts,  $\text{CoSO}_4 \cdot 7\text{H}_2\text{O}$  and  $\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$ , was soluble when 0.01 mM of each salt was added to a 140 g-U/L uranyl sulfate solution, pH 1 at room temperature. When the final concentrations of  $\text{CoSO}_4 \cdot 7\text{H}_2\text{O}$  and  $\text{Na}_2\text{WO}_4 \cdot 2\text{H}_2\text{O}$  were 0.1 mM in the presence of a 140 g-U/L uranyl sulfate solution, pH 1 at room temperature, a flaky precipitate formed within 2 minutes, but precipitation at 60°C did not occur until the concentration of each salt was equal to 10 mM and this precipitate formed immediately. Samples containing 0.01 mM of each salt at room temperature did not form a precipitate after one month. Samples containing 0.01, 0.1, 1, and 3 mM of each salt were kept at 60°C for one month. Precipitation occurred after 1.5 weeks in the samples containing 3 mM of each salt, and after 3 weeks in samples containing 1 mM of each salt. No precipitation was observed in samples containing 0.01 and 0.1 mM of each salt.

Lastly, due to the limited solubility of  $\text{K}_2\text{RuCl}_6$ , precipitation did not occur when the concentration of Ru reached a maximum of 0.5 mM tested in the presence of a 140 g-U/L uranyl sulfate solution at room temperature and 60°C. Samples were kept at room temperature and 60°C for up to one month, and no precipitates formed in either sample.

The potential precipitation of Zr was tested at a concentration about 85 times higher than what is the highest concentration of Zr expected to be present in the SHINE target solution at any point. Even with a very conservative estimate based on data from batch and tracer column studies, at least half of the Zr should remain adsorbed on the titania column. Sn did precipitate when the concentration reached 30 mM at room temperature and 60°C, but that is greater than 200 times the highest expected concentration of Sn. It did not precipitate at a concentration of 20 mM, which is well over 100 times the highest expected concentration of Sn. Ba precipitated when the concentration reached 1 mM independent of temperature, and this is nearly 200 times higher than the highest expected Ba concentration in the target solution.  $\text{CoWO}_4$ , added as two separate metal salts, precipitated at a concentration of 0.1 mM at room temperature, which is only about 10 times higher than the highest Co concentration expected and 1.4 times greater than the highest W concentration expected. Increasing the temperature to 60°C allowed the solids to remain in solution up to a concentration of 10 mM, which is about 900 times higher than the highest concentration expected for Co and about 140 times higher than the highest concentration of W. Precipitation tests using Co and W salts independently showed that both are soluble up to 30 mM at room temperature and 60°C. Ru did not precipitate when the concentration was 0.5 mM,

which is about 65 times higher than the highest expected concentration of Ru in the target solution. The most concerning result is the potential precipitation of  $\text{CoWO}_4$ , which occurs at relatively low concentrations compared to what is expected in the SHINE target solution. Mini-SHINE solutions should provide data more representative of what will be expected in the real SHINE target solution where the effects of a high radiation field will be present.

### *Bubble Experiments*

The purpose of the bubble experiments is to study the thermal hydraulics of the solution volume (a sector of SHINE solution vessel) under the high irradiation field of the electron beam generated by the linac. These experiments will provide valuable information about radiolysis in the solution (especially on bubble formation), thermal dynamics of the solution, and their effects on bubble behavior. Data collected will serve as a validation of computer models which include the thermal hydraulics model created by ANSYS and the nuclear radiation model generated by MCNPX.

To verify and test the experimental designs at the linac, several mini-scale bubble experiments were performed at the 3 MeV Van de Graaff (VDG) accelerator. The experimental setup included a squared quartz tube, sodium bisulfate solution, cooling coils, gas chromatography (GC) system, raster magnets, and two cameras. Different beam currents were applied. Figure 5 shows a picture of the bubbles formed during irradiation at the VDG. Radiolysis (bubble generation), thermal expansion, convection, and radiation damage were observed in the experiments. Photos, videos, and gas formation ( $\text{O}_2 + \text{H}_2$ ) data were recorded. VDG results suggest that the full-scale design of the bubble experiment at the linac is reasonable.

The linac experiments will utilize 20 L of a depleted uranyl-sulfate solution at 140 g-U/L. For the future linac experiments, several important tasks were completed: (1) computer modeling of the experiments, including the thermal hydraulics model and nuclear radiation model, were discussed and established, (2) drawings of the bubble chamber were completed and the whole assembly is being manufactured, (3) the cooling system is under design, and (4) the dump tank is under modification for future storage of the uranyl sulfate solution. The linac beam line has been re-designed, and the big raster magnets to spread the beam were fabricated. The full-scale experiments at the linac will begin in early July.



**Figure 5. Bubble formation and Cherenkov radiation were seen in the mini-scale bubble experiments at the VDG (1.3 M  $\text{NaHSO}_4$  solution,  $\text{pH} = 1.28$ , beam current is  $22\mu\text{A}$ ).**

### **Conclusions**

The mini-SHINE experiments with uranyl sulfate will begin in the next few weeks. A few short irradiations and one production run will be performed before the 1-2 Ci  $\text{Mo-99}$  will be produced

for shipment to Tc-99m generator manufacturer. The solution monitoring and Mo-recovery system is undergoing rigorous inspection for any clogs or hold-ups. Corrosion was observed on the pump head, so a new pump head has been installed for the final sodium bisulfate irradiation. Due to the more corrosive nature of the NaHSO<sub>4</sub> solution, moving to UO<sub>2</sub>SO<sub>4</sub> may resolve low Mo-recoveries.

A “V-3.5” appears to be the best contactor size to process SHINE’s 260-L batches in terms of time and amount of waste generated. Still, this contactor size needs to be tested because specifications for the V-2 and V-5 were utilized to make predictions for the “V-3.5.” The contactors will be used to process the recycled target solution every 4 or more irradiation cycles. The UREX process will generate the largest amount of waste, and a study is underway to optimize the waste streams and prevent formation of GTCC waste.

The effects of a radiation field will dramatically affect the redox chemistry and possibly the pH of the solution slightly. The solubilities of the different elements will change as the redox chemistry and pH of the solution change. A more complete understanding of what will have the greatest potential for precipitation will be identified after the mini-SHINE experiments have been completed using a low-enriched uranium solution as uranyl sulfate.

The bubble experiments at the linac with 20 L of depleted uranyl-sulfate solution will begin in early July 2014.

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