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Research and Development Activities at Savannah River Site's H Canyon Facility

Lindsay Sexton, Kenneth Fuller, Ron Jeffcoat, Chris Armstrong, Tom Severynse
Savannah River National Laboratory, Aiken, SC, USA

Abstract

The Savannah River Site's (SRS) H Canyon Facility is the only large scale, heavily shielded, nuclear chemical separations plant still in operation in the U.S. The facility's operations historically recovered uranium-235 (U-235) and neptunium-237 (Np-237) from aluminum-clad, enriched-uranium fuel tubes from Site nuclear reactors and other domestic and foreign research reactors. Today the facility, in conjunction with HB Line, is working to provide the initial feed material to the Mixed Oxide Facility also located on SRS. Many additional campaigns are also in the planning process. Furthermore, the facility has started to integrate collaborative research and development (R&D) projects into its schedule.

H Canyon can serve as the appropriate testing location for many technologies focused on monitoring the back end of the fuel cycle, due to the nature of the facility and continued operation. H Canyon, in collaboration with the Savannah River National Laboratory (SRNL), has been working with several groups in the DOE complex to conduct testing demonstrations of novel technologies at the facility. The purpose of conducting these demonstrations at H Canyon will be to demonstrate the capabilities of the emerging technologies in an operational environment. This paper will summarize R&D testing activities currently taking place in H Canyon and discuss the possibilities for future collaborations.

Introduction

The Savannah River Site's (SRS) H Canyon Facility is the only large scale, heavily shielded, nuclear chemical separations plant still in operation in the U.S. The facility's operations historically recovered uranium-235 (U-235) and neptunium-237 (Np-237) from aluminum-clad, enriched-uranium fuel tubes from Site nuclear reactors and other domestic and foreign research reactors. Today the facility, in conjunction with HB Line, is working to provide the initial feed material to the Mixed Oxide (MOX) Facility also located on SRS. Many additional campaigns are also in the planning process. Furthermore, the facility has started to integrate collaborative research and development (R&D) projects into its schedule.

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H Canyon Background

H Canyon (Figure 1) was constructed in the early 1950's and began operation in 1955. The facility's operations historically recovered uranium-235 (U-235) and neptunium-237 (Np-237) from aluminum-clad enriched-uranium spent fuel from site nuclear reactors and other domestic and foreign research reactors using a chemical separations process.

Figure 1. H Canyon Facility at Savannah River Site

The H Canyon facility is 1025 feet long, 122 feet wide and 71 feet tall. It consists of both a warm and hot canyon divided into 18 sections/cells with several areas to accommodate the various stages of material stabilization. Between the canyon processing areas are control rooms to monitor equipment and overall operating processes, equipment and piping gallery for solution transport, and sampling aisles for process monitoring (Figure 2). So that worker exposure is minimized, work in the canyon, including maintenance, is remotely performed by unique overhead bridge cranes. Thick, dense concrete walls separate workers from the actual processing areas, providing added protection.[1,2]

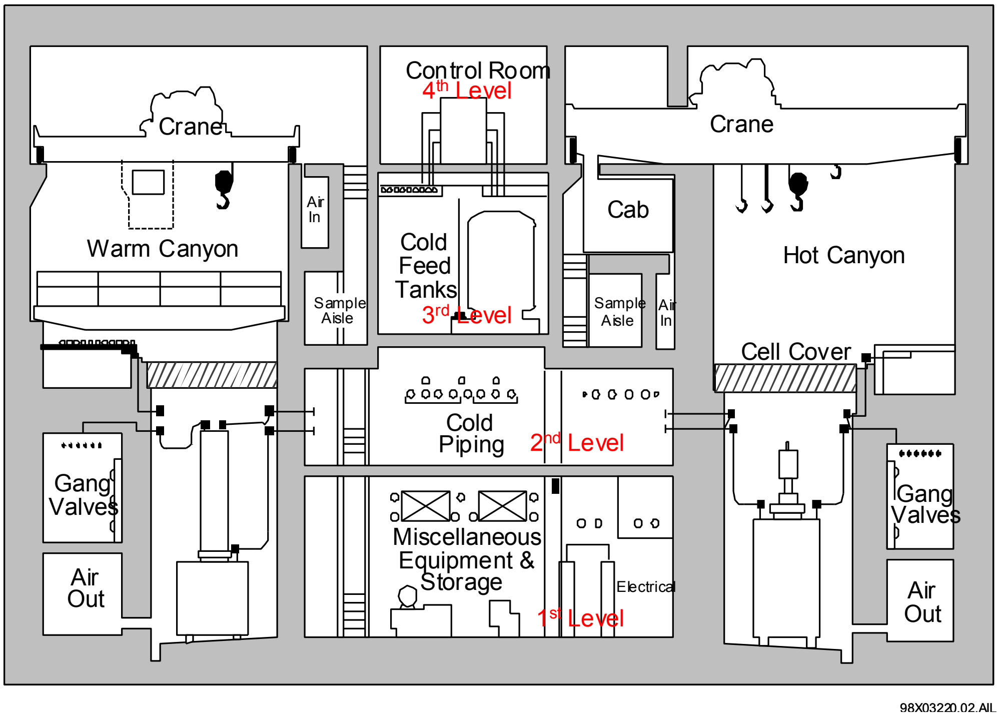

Figure 2. Cross-section of H Canyon.

The sample aisle, located on the 3rd level of both the hot and warm Canyon, is used to obtain sample solutions from Canyon vessels for process control and material accountability. Process solutions are obtained via an air-assisted vacuum lift sampler. The system draws the process liquid from the vessel through piping embedded in the thick concrete wall to a sampling station provided for each vessel, as shown in Figure 3. To ensure homogeneity, the solution in the vessel is agitated for a period of time prior to sampling. The agitated liquid is recirculated through a small vial (15-30 ml) for a minimum of thirty minutes to further ensure homogeneity.

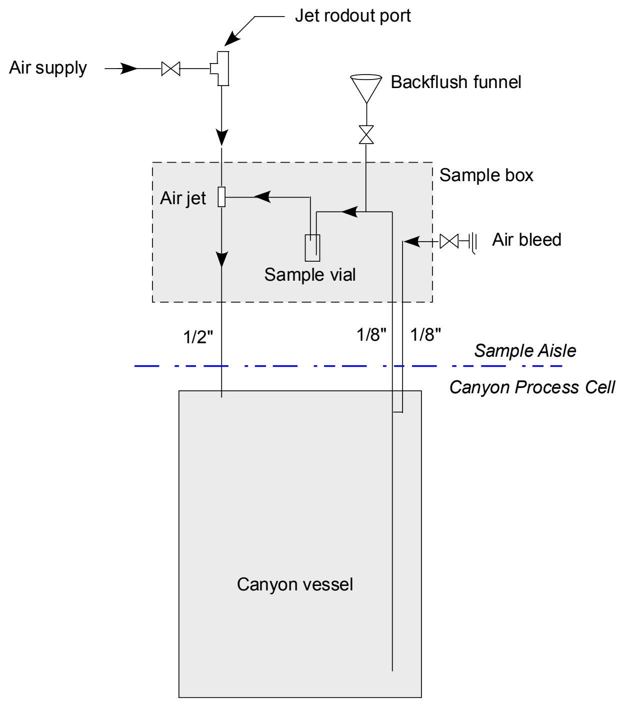

Figure 3. Schematic of H Canyon sampler.

For solutions of low radiation, the sample vials can be placed directly onto the needles to allow circulation of the liquid. For highly radioactive solutions, the sample vial is introduced into the sample box via a heavily shielded "doorstop" made of lead or depleted uranium (Figure 4). The doorstop is rolled into position under the needles, and then raised by a pneumatic cylinder to allow the needles to penetrate the rubber septum in the vial cap.

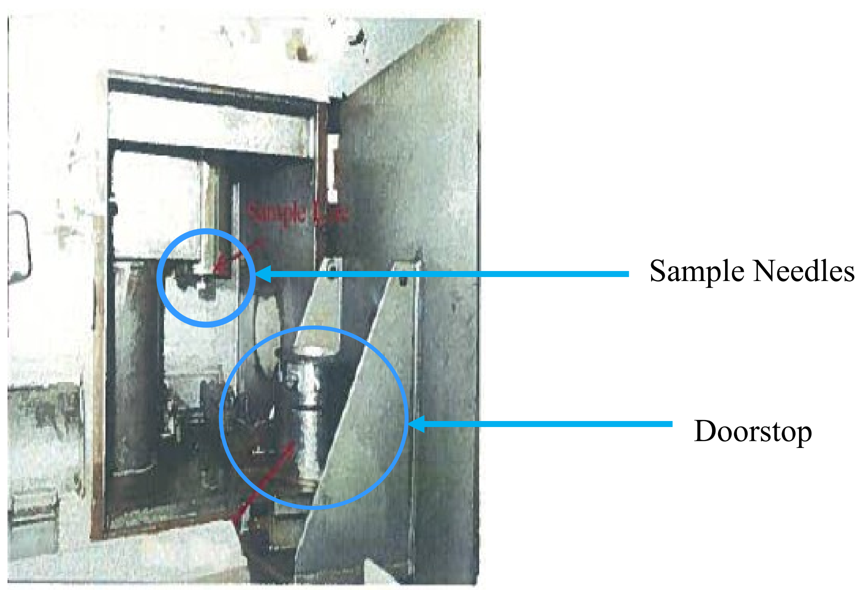

Figure 4. Shielded sampler on hot sample aisle.

The Canyon process for uranium recovery is referred to as the H Modified Process and involves several stages (Figure 5). First of these is dissolving, where the fuel being processed is dissolved in boiling nitric acid mixed with a mercuric nitrate catalyst. Multiple fuel assemblies are packaged in a fuel bundle and multiple fuel bundles are dissolved at a time. After cooling and inspection to ensure the fuel bundles have completely dissolved, the solution is transferred to an accountability tank for sampling. The next stage is the Head End process where the solution from the dissolver is purified and clarified. The solution is fed to an evaporator tank in batches where some of the liquid is allowed to evaporate, concentrating the feed stream. From here, the solution goes to another tank where it is mixed with gelatin to form a slurry. The slurry is then passed to centrifuges where the gelatin, which now contains silica from the dissolved fuel, is separated leaving a concentrated and clarified feed solution.[3]

The next stage is the solvent extraction process which involves mixing the feed solution with either an organic or aqueous solution depending on what needs to be extracted at a particular point. Mixer-settlers are used to carry out these operations. Mixer-settlers flow two streams past each other; one being the feed solution and the other being an aqueous or organic solution depending on what is being removed in that cycle. The two streams mix to some extent as they flow past each other and the desired elements are extracted preferentially into one stream, separating them from the rest of the solution contents. The first cycle extracts uranium and neptunium from the feed solution. This occurs in three multi-stage mixer settler banks. The first extracts uranium and neptunium from plutonium, fission products and other impurities. The next separates the neptunium and uranium and the final stage strips the uranium into a nitric acid solution.[3]

Depending on the desired product from the processing operation, both uranium and neptunium, or plutonium, solutions may be sent to a second cycle for further purification and extraction. The second product cycle involve another series of mixer-settlers that further concentrate and purify the desired elements. The outputs from the second cycle will be pure products. Both high and low activity wastes are generated during the process and each stream has its own cycle for processing and eventual discard as a waste.[3]

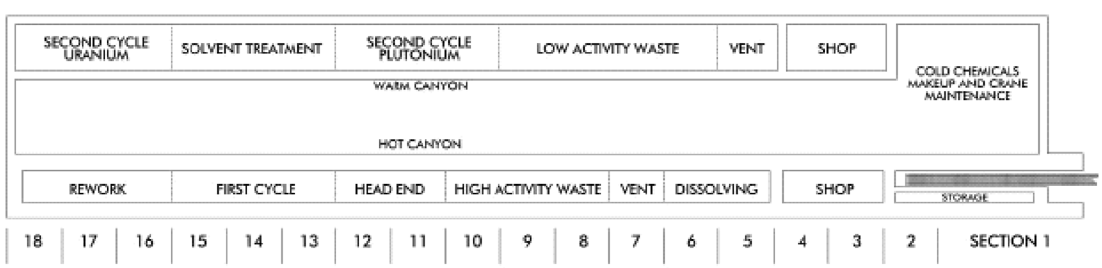

Figure 5. H Canyon processing sections on hot and warm side of Canyon.

H Canyon missions have changed over time, historically being utilized for the Defense Program (U/Pu separation/HEU recovery) as described above. Missions have also supported NASA/military programs for Pu-238 separation for radioisotope thermoelectric generators (RTGs) (i.e., nuclear batteries) and other historical campaigns have involved the recovery of curium, americium, plutonium-242 and thorium. More recently, missions managed by DOE-Environmental Management (EM) have been in support of the cleanup of legacy materials and domestic and foreign research and test reactor fuel recycling. Today the facility, in conjunction with HB Line, is working to provide the initial feed material to the MOX Facility also located on SRS. In addition to this mission, H-Canyon is also processing aluminum-based used nuclear fuel (UNF) from research reactors (foreign and domestic). Many additional campaigns are also in the planning process.

Current R&D Activities

The H Canyon facility has already begun to integrate collaborative R&D projects with current and planned campaigns. The first collaborative R&D project to occur at H Canyon recently took place in March 2013 and involved monitoring the concentration of Pu in MOX feed material. A second R&D project is currently underway which is examining gas effluents from the H Canyon exhaust stack.

UV-Visible Spectroscopy to Monitor Pu Concentration in Solution. SRNL, SRS and Argonne National Laboratory (ANL) have recently completed a demonstration to monitor Pu concentration in an H Canyon process stream using UV-visible spectroscopy. The UV-visible spectroscopy technology is being developed at ANL and sponsored by DOE NNSA Next Generation Safeguards Initiative (NGSI). This project made use of the current H Canyon campaign to create feed material for MOX.

The spectroscopy equipment from ANL was tested in-line with an H Canyon sampler located on the hot side of the Canyon. The processing vessel connected to the sampler contained material directly downstream from the dissolver. The testing location was chosen because previous spectroscopy testing had been completed at the site and the needed fiber optic cables for instrument communication were already in place. These cables ran from the control room (4^{th} level) to the sample aisle (3^{rd} level). Equipment installed in-line with the sampler can be seen in Figure 6. This included an assembly composed of piping connecting a degasser, to prevent air entrainment in the line, and a UV-visible micro-cell to the H Canyon sampling vessel. The assembly was designed to allow for H Canyon to continue collection of destructive analysis (DA) samples from the modified sampler during testing. Additional equipment, including the light source, spectrometer, optical switches, and computer were located in the H Canyon control room.

Figure 6. ANL's UV-visible spectroscopy equipment installed in-line with H Canyon sampler (hot sample aisle).

The UV-vis process monitor development team from ANL spent three days at the H Canyon facility testing their equipment in March 2013. In cooperation with H Canyon and SRNL personnel a great deal was accomplished. The overall result was that the UV-visible process monitor was very effective at detecting the presence of Pu in the process stream. The sampling system was designed, built and installed successfully, and performed well. The overall system operated stably for up to six hours and showed that the Pu concentration did not change throughout that time. The system also detected unexpected metals and forms of Pu, showing that the system is capable of characterizing off normal solution chemistry.

Air Sampling of Gas Effluents. This project is exploiting continuing operation of the H Canyon processing facility at SRS. SRNL will collect and analyze selected gas effluents from H-Canyon operations to answer questions about the efficacy of current and proposed collection approaches. Detailed calculations of the gas content of UNF will provide "ground truth" for collection campaigns involving air. The gases of interest will be collected at the stack, as well as at distance using grab and continuous air samplers. The gas effluents will be compared to the source term calculations, and interpreted in light of meteorological conditions during the collection campaigns. SRNL will deploy collectors at variable distances away from the facilities, consistent with the source terms and expected atmospheric transport. SRNL will also collect background data for the gases of interest during times when processing operations are not occurring.

SRNL has been responsible for obtaining source term information, coordinating with H-Canyon, planning the collection campaigns, preparing the collection system at the stack and at distance, and general deployment of the instrument suite at H-Canyon. SRNL will also provide meteorological fore- and hind-casting support for at-distance measurements.

Gaseous effluents measured have been from batch dissolution processes of mixed UNF bundles consisting of Sodium Reactor Experiment (SRE) and Danish Reactor (DR3) fuels. The proportion of each fuel type in each bundle is tailored to obtain the most effective dissolution process.

The data obtained from this project will inform the nonproliferation community on several questions pertinent to assessing operations at reprocessing facilities. SRNL will assess the performance of current gas concentrating collection methods and we will also look for situations where combining effluent signatures provides additional insight into reprocessing operations.

Potential R&D Activities

Potential Future Nuclear Energy Fuel Cycle R&D. A plan has been developed utilizing H-Canyon to provide for engineering-scale testing of two promising technologies being investigated by the Separations and Waste Forms campaign of the Office of Nuclear Energy's Fuel Cycle Research and Development Program. One technology applies optical and gamma spectroscopic methods to measure actinides in feed, product and waste solutions in a chemical separations process. A second technology provides for characterization and capture of volatile radioisotopes that are generated during fuel irradiation, and released during pretreatment or dissolution of the fuel when it is processed.

These two promising technologies are currently under development at Pacific Northwest National Laboratory (PNNL – optical and gamma spectroscopy) and Oak Ridge National Laboratory (ORNL – capture of volatile radioisotopes). The first includes application of absorbance, Raman, and gamma spectroscopies for measurement of actinides in process solutions. Online or at-line measurement of these materials can provide assurance that processes are operating efficiently to separate products from wastes, and that no materials are being diverted or lost. Application of in-line monitoring during material transfers can provide real-time accountability of nuclear materials. This process monitoring technology would provide the first demonstration of simultaneous measurement of uranium, plutonium, and neptunium at an engineering scale. Integration of the monitoring technology with process operations will inform monitoring instrumentation design in future plants, and allow a cost-benefit comparison of on-line instrumentation with current sampling practices.

A second technology provides for capture of volatile radioactive species (iodine-129, tritium, carbon-14, and krypton-85) that are created during the irradiation of fuel in the reactor. These gases accumulate in the individual fuel rods, and are released when the rods are sheared in preparation for processing. The amounts of these isotopes that are produced are dependent on initial enrichment, fuel burnup, cooling time, and other factors of reactor operation. Capture of these gases is required to meet regulatory requirements for atmospheric releases,[4] and to allow treatment for conversion to a waste form.

Both of these technologies have been successfully demonstrated at the laboratory scale. Operational testing at the plant scale is the next phase required to advance the technology. Using the H Canyon facility at the SRS, operational testing can be performed that will: demonstrate equipment performance at an actual plant scale, assess technology robustness in a high-rad environment, and establish operational requirements and impacts associated with technology deployment.

Planned Future Safeguards R&D. Additional safeguards technology demonstrations are planned for FY14 and beyond. Plans are currently underway to install additional UV-visible spectroscopy monitoring equipment on the warm side of the Canyon. This would be used to monitor the back-end of the MOX feedstock preparation. A long path UV-visible flow cell would be utilized in this case to measure dilute Pu concentrations in filtrate streams.

Discussions are also underway to install and test an x-ray based spectroscopy system being developed at Los Alamos National Laboratory (LANL) in the H Canyon sample aisle. This technology is referred to as Wavelength Dispersive Spectroscopy (WDS) and will be used to measure relative actinide ratios in reprocessing streams.

Work was also recently funded through an Small Business Innovation Research (SBIR) grant with Energy Research Company (Plainfield, NJ) and LANL to explore the use of Laser Induced Breakdown Spectroscopy (LIBS) for in-line measurements of uranium concentrations in processing streams. Discussions and options for testing this technology in the H Canyon sample aisle are currently being explored.

Conclusions and Future Opportunities

The completed and ongoing demonstrations at H Canyon were very successful and have proven the value of collaborative R&D efforts at the Canyon. This work has also opened the possibility for additional projects as added value is seen from both the researchers and facility operators. Over the next several years more projects will be added to the Canyon collaborative R&D portfolio.

Acknowledgments

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