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An Update on the Operational Status of the Savannah River Site H-Canyon

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INTRODUCTION TO H-CANYON

Savannah River Site (SRS), the U.S.'s second large scale nuclear materials production site began construction in 1951. A short history of H-Canyon is in order and has been taken from Ref. 1 and personal interviews with senior staff.

H-Canyon (cross section in Fig. 1) was the second operational radiochemical processing facility on the site, entering service in 1955 processing plutonium. Soon, its sister facility, F-Canyon which entered service in 1954 became the primary plutonium workhorse and H-Canyon moved to a primary mission of recovering highly enriched uranium (HEU) from used reactor fuel from the SRS reactors. Of the five Department of Energy separations canyons (3 at Hanford and 2 at SRS), it is the only one remaining in operation. Over the decades H-Canyon has had a variety of missions supporting national defense, research, space exploration, and medical and industrial uses of radioisotopes.

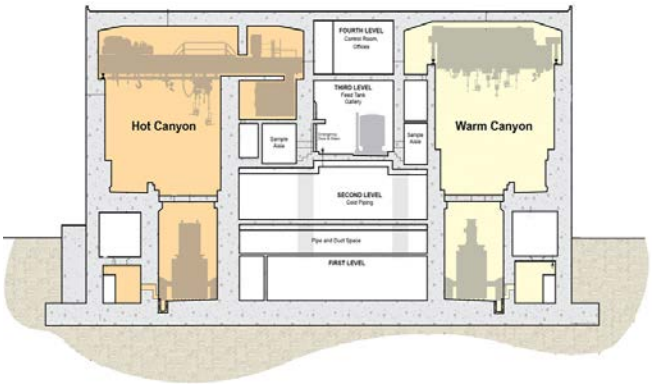

Fig. 1: H-Canyon cross section.

In addition to reprocessing SRS reactor fuels, H-Canyon recovered HEU from numerous research reactors throughout the U.S. and world – a mission it continues today. It has recovered numerous specialty isotopes in industrial quantities including Pu-240, Np-237 for Pu-238 production, Pu-238 for space exploration, Am-241 and Am-243, and various isotopes of curium. Supplies of SRS americium and curium are still being used in isotope production today. Np-237 was converted to oxide targets for irradiation to Pu-238. Pu-238 production ran from 1961-1984 (concurrent with Np-237 recovery) and from 1992-1995. From 1964-1969, H-Canyon ran the THROEX process to process irradiated thorium targets, providing a significant contribution to the nation's storehouse of U-233. From 1969-1980, the canyon operated a novel production scale electrolytic dissolver cable of reprocessing stainless steel and zirconium clad fuels. In 1997 the restart program began which repurposed the facility to a mission of stabilization, disposition, and reuse of nuclear material. Legacy plutonium was stabilized by 2002, and legacy neptunium by 2008. The last of the unirradiated SRS HEU reactor fuel was dispositioned by 2006. That material became the first downblend batch, producing fuel for the Tennessee Valley Authority power reactors. Specialty HEU materials from the NNSA were processed in 2008-2010. Plutonium was dissolved, purified, and converted to oxide for the MOX campaign from 2014-2017. Today, other missions continue.

CURRENT MISSION

Overview

At the time of this writing, Savannah River Site's H-Canyon is considered to be a fully operational facility. This means that the facility has a mission and that all necessary processes are in place and operating in order to fulfill that mission. The primary mission is to disposition HEU material, namely from foreign and domestic research reactors. The uranium is separated from its irradiated fuel matrix, purified, and then stored to be downblended for commercial power reactors or other low enriched uranium (LEU, <20 wt.% U-235) applications. Fig. 2 shows the basic process flow. Much of the discussion below is adapted from Ref. 2 and supplemented by the author's personal experience as a principle engineer for the facility.

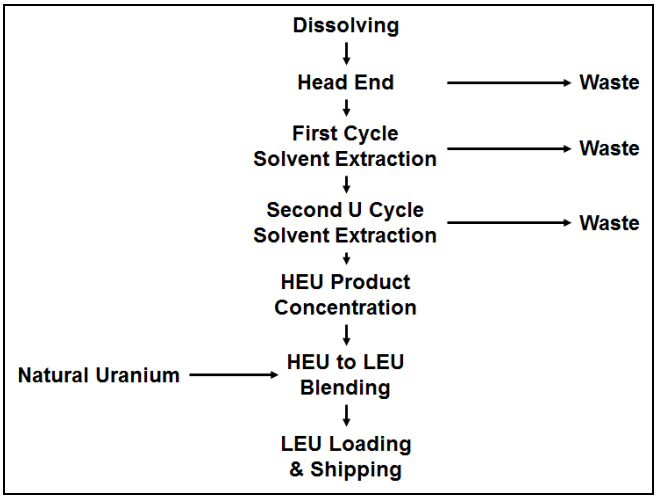

Fig. 2: Process flow diagram.

Feed Material

H-Canyon currently receives three primary HEU feed streams differentiated by geometry and chemical form.

The most prominent and diverse feed stream is used nuclear fuel from research reactors. This fuel is irradiated uranium-aluminum alloy in aluminum cladding. The assemblies come from a variety of domestic and foreign (now 29 countries) research reactors and typically measure a few inches in radial geometry and two to four feet in active fuel length (Fig. 3). Most fuels had beginning of life enrichment >90 wt.% U-235, but some were ~20 wt.% fuel. The irradiation histories vary such that for criticality safety purposes fresh isotopics are assumed. These used fuel assemblies are stacked two to four into ~14-foot aluminum tubes called bundles and the bundles, along with the fuel, are dissolved in nitric acid.

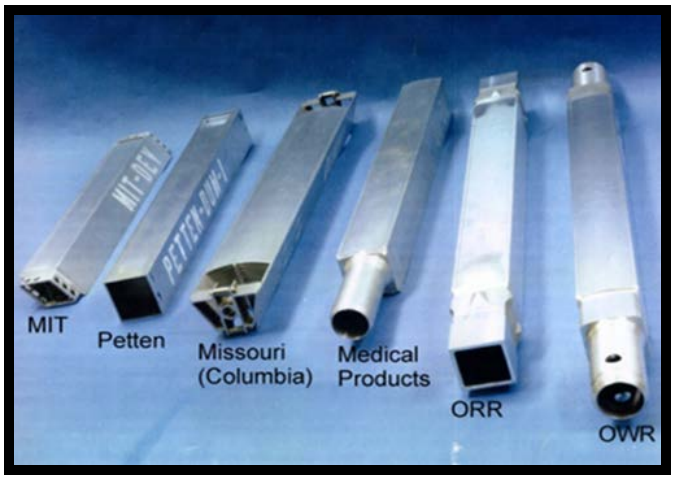

Fig. 3:Assorted research reactor fuel assemblies.

A second feed stream is the Oak Ridge National Laboratory High Flux Isotope Reactor cores (Fig. 4 and 5). These cores arrive in two components which are a unique geometry. They are irradiated uranium oxide (U_3O_8) in aluminum cladding which are dissolved in a dissolver insert especially designed for them. HFIR's high aluminum content combined with the nitric acid dissolving chemistry makes criticality in the dissolver not credible, expediting this processing stream.

The third stream is called target residue material. This is a dissolved uranyl nitrate solution received from off-site. HEU was alloyed with molybdenum and then irradiated to produce Mo-99/Tc-99. The uranium-molybdenum alloy targets were dissolved off-site to recover the molybdenum and its technetium daughter, mainly for medical uses. The residue HEU uranyl nitrate solution is returned to SRS for disposition.

The Savannah River Site continues to receive used nuclear fuel from research and test reactors. Some of this fuel is in unique one-of-a-kind geometries which do not yet have a specified disposition path.

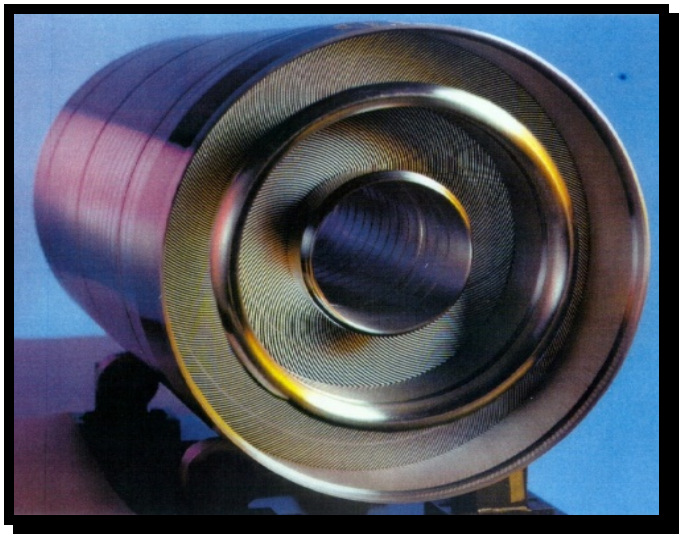
Fig. 4: High flux isotope cores.

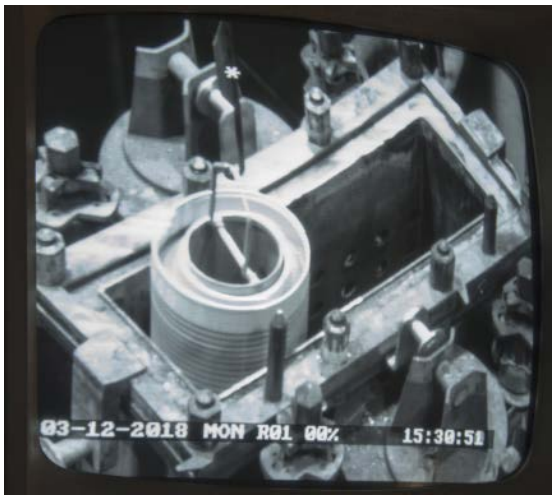

Fig. 5: Remote video feed of used HFIR core being dispositioned in a dissolver on March 12, 2018.

Clarification, Separation, and Purification

At its core H-Canyon has been, is, and always will be an industrial scale radiochemical separations facility. The three feed streams are blended together to achieve an enrichment mix less than 73 wt.% U-235 to meet chemistry flowsheet requirements. The solution is then combined with gelatin, heated, and centrifuged, in a process called a strike. This operation is to remove silica which can create an emulsion that inhibits separation and purification during solvent extraction. When the facility processed short-cooled reactor fuels, a potassium permanganate strike process to remove zirconium, niobium, and ruthenium fission products was also performed. These isotopes have decayed away in the current fuel and that strike is no longer needed. The centrifuging also removes any undissolved fission products

and cladding fragments; uranium losses are negligible. The clarified solution is adjusted with acid and if necessary reduced in enrichment using fresh uranyl nitrate solution of natural uranium.

The solution is then sent to a counter current solvent extraction system with three banks of mixer settlers comprising 36 stages of counter-current processing. The acidic aqueous solution is contacted with a 7.5 vol.% tributyl phosphate (TBP) solution that is dissolved in an n-paraffin diluent. In this system, called First Uranium Cycle, fission products are rejected through control of valence states as high activity waste in the first bank. The trace amounts of neptunium, plutonium, and other transuranics are rejected through control of valence states as low activity waste in the second bank. In the third bank, the uranium is transferred from the organic back into the aqueous phase and a relatively pure product is removed from the system.

The aqueous solution is then acid adjusted and re-concentrated before being fed to a second solvent extraction system, known as Second Uranium Cycle. This process also uses the same mixture of TBP-diluent and the principle of valance state control to strip out any remaining transuranics in the first 16-stage bank. The second 12-stage bank transfers the now highly purified uranium back into the aqueous phase to produce the final product. This final solution is then adjusted, and concentrated, and stored until ready for downblend. The Second Uranium Cycle may also be used for polishing of product solution to meet the purity standards necessary for commercial reactor fuel.

Waste Disposal and Chemical Recycling

The facility has five fully operational systems to disposition waste and recycle chemicals.

The High Activity Waste (HAW) system receives mainly fission product bearing aqueous acid solution wastes from the centrifuge and the First Uranium Cycle. Dilute aqueous HAW is concentrated and the acid overheads processed for reclamation of the nitric acid. The slurry of HAW from the centrifuge is combined with the concentrated aqueous solution downstream. The wastes are neutralized to a caustic pH. The caustic protects the integrity of the waste storage tanks to which the facility wastes are sent. The Low Activity Waste (LAW) system works similarly except that it receives the transuranic wastes from First and Second Uranium Cycles, which are typically low activity, low concertation of plutonium and neptunium.

The TBP n-paraffin solvent is also recycled. Each separations cycle has its own dedicated solvent washer in the Solvent Recovery system. The solvent is washed with alkaline sodium carbonate solution to remove degradation products mainly generated by radiolysis. The wash also neutralizes the trace entrained acid which is sent out to waste.

The acid solution recovered from solvent washing and small samples returned from laboratory analysis are collected in the Sumps, Solvent, and Sample Return (SSSR) system. SSSR also processes the canyon's sumps. H-Canyon is divided into eighteen sections of processing equipment and each section has a sump. The sump collects any spilled solution and allows it to be transferred out of the cell. Like HAW and LAW, SSSR wastes are concentrated, the acid recovered, the waste neutralized, and finally sent to the tank farm for ultimate disposal.

The acid overheads collected in the above four systems are purified and concentrated in the Acid Recovery Unit, which is a standard piece of industrial equipment that purifies the acid and re-concentrates it back to 64 wt.% HNO_3.

Downblending Operations

The process of downblending involves combining HEU uranyl nitrate solution purified by H-Canyon processes with clean, fresh natural uranium uranyl nitrate solution. The blend is adjusted such that the effective enrichment (including negative reactivity of trace materials) of the combined solution is LEU. Since downblend operations began in 2003, this isotopic target has been 4.95 wt.%. The uranyl nitrate solution is shipped off-site to a fuel fabricator which converts it to uranium dioxide pellets for use as commercial reactor fuel (Fig. 6).

The downblend operations are carried out in massive tanks outside of the canyon in what is called H Outside Facilities which is for cold chemical and very low radioactivity processes. The process is currently in standby as another large batch of uranyl nitrate is accumulated from canyon operations.

Fig. 6: Purified and downblended LEU solution being shipped off-site for use in commercial reactor fuel fabrication.

RECENT IMPROVEMENTS

Above and beyond the normal maintenance repairs of equipment, over the last five years there have been a number of improvements to the H-Canyon facility aimed at

improving efficiency, bringing in new technology, and facilitating current missions. These include:

- Replacement of the vessel in which dissolution of research reactor fuel is carried out
- Installation of advanced spectrophotometers on several process vessels to replace conductivity meters and uranium colorimeters. This one device reads uranium and acid concentration in real time replacing two legacy systems.
- Installation of mechanical pumping systems with passive safety designs to receive off-site HEU uranyl nitrate solution
- Unused evaporator has been returned to service in the low activity waste system to improve efficiency
- A passively safe vacuum has been developed to clean out the sumps

However, one of the most important improvements is the training and qualification of a new generation of shift managers, operators, building technicians, and system engineers. This ensures this national asset keeps running safely and efficiently for years to come.

FUTURE MISSIONS

The future of H-Canyon still looks bright and the U.S. will continue to have a fully operational shielded radiochemical separations facility for years, potentially decades, to come. Liquid HEU receipts will continue until 2020 (the same year H-Canyon turns 65). The current DOE record of decision (Ref. 3) on processing specifies 1,000 research reactor fuel bundles and 200 HFIR cores (120 from SRS and 80 from Oak Ridge National Lab) through 2026. However, the site has now received and is storing over 8,900 research reactor assemblies totaling more than 3,000 bundles. Meanwhile, used HFIR cores continue to be produced at Oak Ridge National Lab. To fully disposition all the material will require operations into the late 2030s or early 2040s.

The facility continues to be evaluated for new and exciting missions. Downblending operations, currently specified for 4.95 wt.% equivalent enrichment, are being evaluated as a potential source for 19.75 wt.% uranium, to become one of DOE's high assay LEU sources.

The facility has been selected as the disposition path for stainless steel clad plutonium research reactor plates. This mission will refurbish and restart the electrolytic dissolver. This mission returns the capability to process stainless steel and zirconium clad and zirconium alloyed uranium and plutonium fuels.

Scoping work is being down done to provide a disposition path for some DOE/NNSA materials that currently have no disposition path. Some of these include disposition of unique one-of-a-kind uranium fuels and test reactor cores. This also includes high temperature gas reactor pebble fuels (approximately 1,000,000 of them). While the fissile material content of the pebbles (mixed HEU, U-233, and depleted thorium) is similar to other materials the facility has processed in the past, they would require new equipment running a novel non-aqueous digestion process – a first for the facility. It is important to note that the DOE has not made any final decision on missions still under scoping.

ACKNOWLEDGEMENTS

This work is in no small part the result of interviews with H-Canyon personnel, whose hard work, dedication, and pride in their facility are evidenced by the longevity and versatility of this now unique national asset. Special thanks are due to Michael Lewczyk, Virginia Magoulas, Maxcine Maxted, and Charles Georgen (ret.).

REFERENCES

1. M. B. Reed, "*Bringing it to Form: A Thematic Study of Savannah River Site's Separations Processes, F and H Areas*", New South Associates, (2013)
2. "*H-Canyon and Outside Facilities, H-Area Documented Safety Analysis*", S-DSA-H-00001, Rev. 12, Savannah River Nuclear Solutions (2017)
3. "*Savannah River Site Spent Nuclear Fuel Management Final Environmental Impact Statement*", Aiken, SC (DOE/EIS-0279, 2000; SRS SNF EIS, Amended April 2013 EIS-0279)