

Recovery and Blend-Down Uranium for Beneficial use in Commercial Reactors – 13373

Virginia Magoulas
Savannah River National Laboratory, Savannah River Site, Aiken, SC 29808,
virginia.magoulas@srs.gov

ABSTRACT

In April 2001 the Department of Energy (DOE) and the Tennessee Valley Authority (TVA) signed an Interagency Agreement to transfer approximately 33 MT of off-specification (off-spec) highly enriched uranium (HEU) from DOE to TVA for conversion to commercial reactor fuel. Since that time additional surplus off-spec HEU material has been added to the program, making the total approximately 46 MT off-spec HEU. The disposition path for approximately half (23 MT) of this 46 MT of surplus HEU material, was down blending through the H-canyon facility at the Savannah River Site (SRS). The HEU is purified through the H-canyon processes, and then blended with natural uranium (NU) to form low enriched uranium (LEU) solution with a 4.95% U-235 isotopic content. This material was then transported to a TVA subcontractor who converted the solution to uranium oxide and then fabricated into commercial light water reactor (LWR) fuel. This fuel is now powering TVA reactors and supplying electricity to approximately 1 million households in the TVA region.

There is still in excess of approximately 10 to 14 MT of off-spec HEU throughout the DOE complex or future foreign and domestic research reactor returns that could be recovered and down blended for use in either currently designed light water reactors, ~5% enriched LEU, or be made available for use in subsequent advanced “fast” reactor fuel designs, ~19% LEU.

INTRODUCTION

In 1991, the Cold War ended and operation of the SRS production reactors ceased, leaving highly enriched uranium (HEU) materials without a disposition path. In 1995 the President declared approximately 200 metric tons (MT) of fissile material surplus to national security needs. The Department of Energy (DOE) created the National Nuclear Security Administration (NNSA) Office of Fissile Materials Disposition (OFMD) to demonstrate the nation’s commitment to non-proliferation and to manage the disposition of these surplus materials. The HEU Disposition Program Office (HDPO), located at Y-12 National Security Complex, was established to plan, integrate, and manage surplus HEU disposition activities across the DOE complex. The purpose of the HEU disposition program is to make the surplus HEU unusable for weapons, to dispose of it in a safe, secure, and environmentally acceptable manner, and to recover the economic value to the maximum extent possible. Due to the huge amount of recoverable energy stored in the HEU, and its economic value, HDPO planned to convert approximately 85% (over 155 MT) of the available HEU to commercial or research reactor fuel. The remaining 15% of the surplus HEU would not suitable for use as commercial fuel and will be disposed of as waste. Approximately 174 metric tons (MT) of the 200 MT declared surplus was HEU, which was broken down into two major categories; on-specification (on-spec), materials capable of meeting the American Society for Testing and Materials (ASTM) commercial nuclear fuel specifications after down blending; and off-specification (off-spec)

materials, which when down blended would not meet the ASTM specification for normal commercial nuclear fuel. Approximately 60 metric tons of this surplus HEU inventory is considered off-spec. Within the HDPO, the Off-Spec Fuel Program was formed to focus on the disposition of this off-spec material. Approximately 23MT of the 60 MT off-spec surplus uranium materials, originated as legacy materials at the Savannah River Site (SRS). The SRS is a DOE facility located in South Carolina on the Savannah River near Augusta, Georgia.

In the 1980's, prior to the end of the Cold War, SRS operated four heavy water moderated, production reactors with associated spent fuel basins, two chemical reprocessing facilities, and a fuel fabrication facility. The chemical processing facilities or "canyons" dissolved the fuel, separated and purified the HEU from high level waste constituents. The Y-12 Plant at Oak Ridge, Tennessee received HEU solutions from SRS, performed solvent extraction purification and converted the material to uranium metal (buttons). The buttons were returned to SRS where they were mixed with clean 93% U-235 material to replenish material consumed by reactor operation and then fabricated into new fuel for the SRS reactors. When the Cold War ended, this process was stopped, resulting in surplus off-spec uranium being located at SRS without a disposition path.

Alternatives were evaluated for the disposition of this material. The primary ones were: (1) down blending HEU to LEU for use as commercial fuel, and (2) down blending HEU to LEU and discarding as waste. Both pathways met the non-proliferation and permanent disposition goals, but analyses indicated that use in a commercial power reactor would be much less expensive and had the potential to generate significant cost savings as opposed to the blend down to waste alternative.

In 1994, DOE published an Expression of Interest (EOI) to the nuclear industry regarding the potential use of this material. TVA was the sole respondent to the EOI. Several conceptual studies were also conducted to simulate performance of off-spec fuel in commercial power reactors (reference 1). The results of simulated performance testing of off-spec fuel showed that by increasing the U-235 content in the fuel the neutron absorption property of the ^{236}U can be overcome without impacts to reactor safety. An Environmental Impact Statement was performed in 1996 whose outcome stated: "...to eliminate the proliferation threat of surplus HEU by blending it down to low enriched uranium (LEU), which is not weapons-usable. ... The Preferred Alternative is, where practical, to blend the material for sale as LEU and use over time, in commercial nuclear reactor fuel to recover its economic value." A study of the alternatives showed a significant economic benefit to the government when used as fuel (reference 2). In 1996, the Interim Management of Nuclear Materials (IMNM) Supplemental Record of Decision was issued allowing the HEU derived from the legacy SRS HEU solutions and SRS fuel to be down blended and used in nuclear power reactor fuel. At this point, the DOE - Environmental Management (DOE-EM), National Nuclear Security Administration (NNSA), and the TVA initiated formal negotiations to develop a program to produce this nuclear power reactor fuel. This program is the Off-Spec Fuel Program. While NNSA manages the Off-Spec Program, DOE-EM manages the facilities at SRS.

USE OF SRS INFRASTRUCTURE AND FACILITIES

As previously stated, approximately 30 MT of the off-spec HEU is slated to be processed through SRS H Canyon. While the initial 18 MT of HEU was legacy material stored at SRS; the subsequent approximately 12 MT are materials from the deinventory of other DOE facilities and receipts of both foreign and domestic research reactor returns. The SRS has an infrastructure to support receipt, and storage of excess uranium materials in both L-area and K-area storage facilities. SRS has the capabilities to transport the uranium material from L or K-area storage facilities to H canyon, the only nuclear chemical separations facility operating in the US. After the highly enriched uranium (HEU) is separated and purified in H Canyon, it is down blended to low enriched uranium (LEU) in H Outside Facilities (HOF) using natural uranium (NU) solutions provided by the end user.

H Canyon Background

H Canyon was constructed in the early 1950s and began operation in 1955. The facility's operations historically recovered U-235 and 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.

The H Canyon facility is 312 meters long, 37 meters wide and 21.5 meters 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 galleries for solution transport, and sampling aisles for process monitoring (Figure 1). 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.

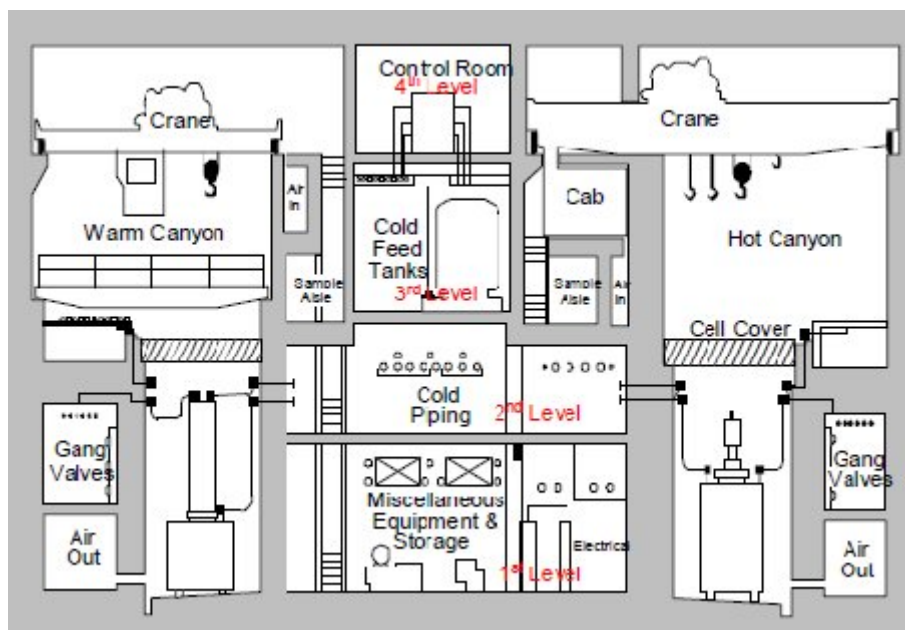


Figure 1. Cross-section of H Canyon

The H Canyon process is a modified PUREX process (i.e., H Modified Process) that involves several stages (Figure 2). 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 evaporated, concentrating the feed stream. From here, the solution goes to another tank where it is mixed with gelatin to form a slurry mixture. 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.

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-settler banks 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. 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.

Depending on the desired product from the processing operation, both uranium or neptunium solutions may be sent to a second cycle for further purification and extraction. The second product cycles involve another series of mixer-settlers that further concentrate and purify the

desired elements. The outputs from the second cycles will be pure product. Both high and low activity wastes are generated during the process and each stream has its own cycle for processing and eventual discard as waste.

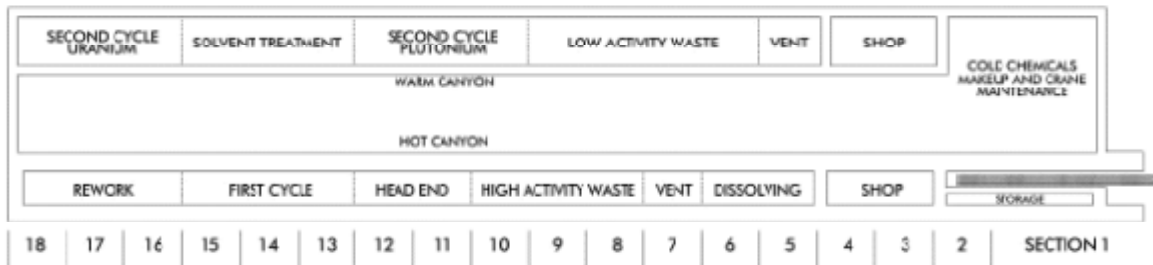


Figure 2. H Canyon processing sections on hot and warm side of canyon

SRS Down Blending Process

Once the HEU material has been dissolved and purified in H canyon, the HEU nitrate solution is transferred to HOF for down blending (Figure 3). HOF is located behind the H Canyon facility.

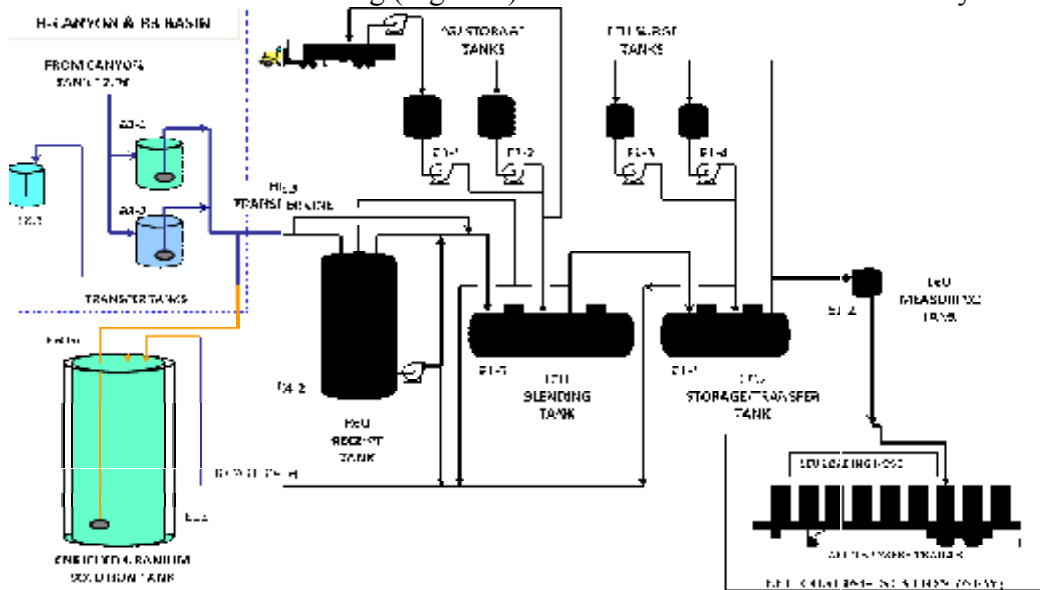


Figure 3. HOF Down Blending Process

The blend grade HEU solution is received in one of two tanks where it is sampled to ensure the quality of the uranium product from H canyon. After meeting the HEU specification for blend down, the material is transferred to the HEU receipt tank. Once the quantity of HEU needed for down blending has been received, a subsequent sample of the material is taken to ensure the proper blend ratio is set. For down blending, the HEU is mixed with natural uranium (NU) which is received in a tanker truck as a uranyl nitrate solution. The HEU and NU are then blended in the LEU blending tank. After blending, the mixture is sampled and verified to meet

the LEU specification for fuel fabrication. The LEU blend batch is then transferred to the storage/transfer tank. SRS does not currently have the capability to convert the uranium solution to oxide and then fabricate the fuel. Therefore, the blended LEU solution is transported elsewhere for the conversion process and fuel fabrication. HA-Line is equipped with a station for loading the blended LEU solution into Liquid-Rad (LR230) type B shipping containers to support this transport.

THE OFF-SPEC FUEL PROGRAM

Since 2003, SRS has down blended approximately 23 MT of HEU producing approximately 301 MT of 4.95% U-235 LEU. This blended LEU (BLEU) solution was part of the Off-Spec Fuel Program managed by NNSA.

The Off-Spec Fuel program began when DOE and TVA signed a Memorandum of Understanding (MOU) to evaluate the potential use of off-spec HEU material for fuel in the TVA reactors. The TVA Sequoyah Nuclear Plant reactors were the initial choice for use of this off-spec material. A 1997 agreement provided some of the candidate HEU material to TVA for the production of four lead unit assemblies (LUA) for irradiation in TVA's Sequoyah Unit #2 reactor. TVA developed partnerships with Nuclear Fuel Services (NFS) and AREVA to produce these lead test assemblies. The blended LEU fuel assemblies were fabricated, and loaded into the TVA Sequoyah Unit #2 in 1998 where they performed as predicted and without incident.

With the success of the lead test assemblies, TVA and DOE signed an Interagency Agreement (IA) in April of 2001 to disposition off-spec HEU materials. Initially approximately 33 MT of surplus off-spec HEU was sufficiently characterized to be allocated to this agreement. The IA contains a provision which allows for the addition of new HEU/LEU material to the Off-Spec Fuel Project, if both parties agree. Subsequent additions have been made bringing the current total to approximately 46 MT of the possible 60 MT of off-spec HEU.

The agreement stated the material would be blended down to a 4.95% ^{U-235} LEU solution and converted to powder and used as off-specification TVA commercial nuclear fuel. The resulting fuel was initially slated to be used in the Sequoyah pressurized water reactors (PWR) but at that time, those reactors were deemed necessary for tritium production, so the fuel use was changed to the Browns Ferry Nuclear Station boiling water reactors (BWR).

An LEU acceptance specification was developed based on the ASTM specification for normal fuel. Small deviations were made to accommodate the off-spec material characteristics. Levels of neptunium and plutonium were required to be extremely low for personnel exposure protection, waste handling, and NRC licensing as non-TRU facilities. NFS, located in Erwin, Tennessee, and the SRS are the two facilities within the off-spec program designated for down blending HEU to LEU to meet the LEU acceptance specification.

Approximately 23 MTU of the off-spec HEU had levels low enough to meet the NFS facility permit requirements and the program plans called for this material to be shipped from throughout the DOE complex (but primarily at SRS, Y-12, and Idaho National Laboratory (INL)) to NFS for down blending. The NFS facility permit limits would not allow down blending of the remaining

off-spec HEU materials originally in inventory at SRS due to the elevated levels of ^{232}U , ^{234}U , and ^{236}U . These contaminants as well as various levels of transuranic actinides were due to numerous cycles through the reactor and recovery process. The SRS inventory included irradiated fuel, unirradiated fuel, solutions in various stages of purification, ingots of recast fresh fuel, and metal buttons.

Originally the proposal for SRS down blending was to use depleted uranium (DU) already at SRS for blending the HEU to LEU. Since the SRS canyon's primary criticality control is concentration, the solvent extraction process does not run at saturation. This results in a relatively high level of impurities and the isotopic diluent purity is critical to dilution of any HEU solution impurities to low levels. The SRS DU was not pure enough to be used because the resultant LEU solution would not be within the down blended LEU specification. Use of natural uranium (NU) provided the maximum impurity dilution and minimum expense. In 2003, SRS completed H Canyon facility modifications, and began processing the SRS legacy spent fuel and HEU solutions in preparations for down blending.

The down blended LEU solutions are sent to AREVA. A LEU loading facility was constructed at SRS, to allow for loading and shipping of the solutions to AREVA. This facility allows for the SRS LEU solution to be loaded into a DOT Type B shipping container known as the Liqui-Rad LR 230 container. This container was designed and licensed by Columbiana Hi-Tec specifically for the off-spec program. TVA paid for the design and licensing of the container and AREVA designed and purchased five trailers each outfitted with nine of the LR 230 containers to make the deliveries from the SRS facilities to the AREVA facilities. AREVA receives the LEU solutions and converts them to oxide. The oxide power is stored until feed material is requested from the fuel fabrication facility. The fuel fabrication facility presses the oxide into fuel pellets. These pellets are loaded into fuel rods which form the fuel assemblies know as blended low enriched uranium (BLEU) fuel. The BLEU fuel assemblies are shipped in casks to either TVA's Brown's Ferry Nuclear Plant for use in Units #1, #2 and #3 or Sequoyah Unit #2.

THE OFF-SPEC FUEL PROGRAM "BY THE NUMBERS"

Initially approximately 33 MT of surplus off-spec HEU was sufficiently characterized to be allocated to this program through the DOE/TVA agreement. Based on the agreement from both parties, approximately 6 MTU of additional surplus off-spec HEU was added in April 2004 and another 1 MT was added in a 2005 modification. These additions brought the total off-spec HEU allocated to the Off-Spec Fuel Project to approximately 40 MTU. In 2009, provisions were made to the agreement to add another approximate 20MT of HEU, with only a guarantee to TVA of approximate 5.6MT. This additional ~20MT is made up of ~7MT of surplus off-spec HEU from the various NNSA laboratory inventories, and ~13MT of spent nuclear fuel from foreign research reactors (FRR) and domestic research reactors (DRR). This brings the total of surplus HEU for down blending disposition to ~60MT.

To date the program has completed down blending all the material currently in the program (approximately 46 MT of HEU) resulting in the production of approximately 613MT of LEU solutions delivered for oxide conversion. The 46 MT dispositioned through this program has created a significant reduction in the storage of legacy special nuclear material (SNM) across the

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DOE complex. It has allowed Y-12 to de-inventory, consolidate, and meet DNFSB commitments. It has dispositioned material allowing Lawrence Livermore National Laboratory (LLNL) to stay on track to deinventory below SNM Category II by the end of 2012. It has removed all the SRS legacy spent fuel and legacy HEU solutions that were being stored at the SRS. It has also removed materials from INL (meeting a DNFSB commitment) and Los Alamos National Laboratory (LANL) which had no other disposition path. By this program allowing these facilities to disposition their legacy materials it also allows for reduction in the safeguards and security requirements, eliminates the strain on storage capacity, and eliminates the storage costs for these materials.

There is a pending decision whether to process the additional approximate 10 to 14MT of surplus HEU, which is mainly spent nuclear fuel, through the H-canyon facility at SRS or to establish a way to continue to store this material indefinitely. While safe storage is possible, it does not eliminate the risk of proliferation of this material. Safeguards and security measures are still required. With the down blending process, this material is made non-proliferable therefore reducing the attractiveness to an adversary. This program has already eliminated more than 1,000 nuclear warhead equivalents.

This program transforms the weapons-usable special nuclear material into a form that provides an economic benefit - electricity for the people served by TVA. Making valuable use of this surplus material as opposed to storing it waiting for the next generation to resolve.

The current DOE/TVA program will provide a return to the U.S. Treasury Department of an estimated \$230 million by the end of 2016 (reference 3). Down blending of the additional ~10 to 14 MT as part of the DOE/TVA program, will continue to provide returns (estimated at an additional \$300 -400 million depending on the market) to the U.S. Treasury Department while eliminating the cost of long term material storage.

CONCLUSION

There is still in excess of 10 MT of off-spec HEU throughout the DOE complex or future foreign and domestic research reactor returns that could be recovered and down blended for use in either currently designed light water reactors, ~5% enriched LEU and put into the DOE/TVA Off-Spec HEU Program, or be made available for use in subsequent advanced “fast” reactor fuel designs, ~19% LEU.

Either way this material left from the Cold War can be given an important peaceful use – energy. Down blending of the HEU material continues to be in line with the U.S. commitment to reduce potential nuclear proliferation by reducing the amount of stored weapons-usable material that is excess to the national security needs. It also has eliminated the environmental concerns associated with this material.

In 1953, almost 60 years ago, then President Dwight D. Eisenhower spoke before the United Nations calling for the peaceful use of atomic energy. His vision that day was to

“seek more than the mere reduction or elimination of atomic materials for military purposes. It is not enough to take this weapon out of the hands of soldiers. It must be put into the hands of those who will know how to strip its military casing and adapt it to the arts of peace.”

Not only does down blending surplus HEU support President Eisenhower’s vision in the reduction of surplus nuclear material, but it has also adapted the material for process and delivery of a product that resulted in clean and affordable electricity through nuclear power. The many benefits of down blending include:

- Significantly reduces the storage of material
- Reduces Safeguards and Security requirements
- Addresses DNFSB concerns about storage of material
- Reduces the tremendous strain on storage capacity
- Reduces U.S. stockpile of weapons-usable fissile materials
- Reduces Costs to taxpayers (storage, security, disposal)
- Provides a vehicle for deinventory of multiple DOE Sites (SRS, Y-12, ETTP, INL, LLNL, LANL)
- Provides jobs
- Allows the U.S. to maintain workers skilled in the disposition of special nuclear materials
- Provides affordable electricity

Conversion of the excess HEU materials to a useable fuel can be a “win-win” for all parties involved. The material owner, DOE, reduces the footprint requiring safeguards and security of various facilities, and allows for the elimination of long term material storage and resulting costs; and whether the end user wants an ~19% LEU fuel in attempts to advance a fast reactor program or wants to continue to fabricate ~5% LEU fuel, it is using a surplus resource that produces the economic benefit of lower-cost energy for the people and makes the world a safer place. To date, the LEU already produced and shipped is equivalent to approximately 206,270,890 barrels of crude oil, which produces enough power to generate electricity for feed approximately 10 million U. S. households for an entire year.

- KWh generated per unit of fuel used:
 - 2,141 kWh per MT of Coal or 2.141 kWh per kg of Coal
 - 350 kWh per 100 L of Natural Gas
 - 610 kWh per Barrel of Residual Fuel Oil, or 3.8 kWh per Liter

REFERENCES

1. D. Biswas and W. Bickford, “Potential Utilization of SRS Uranium with High ²³⁶U Content as Commercial LWR Fuel,” Transactions of the American Nuclear Society, Volume 72, 1995
2. U.S. Department of Energy, Disposition of Surplus Highly Enriched Uranium, Final Environmental Impact Statement, Volumes I & II, June 1996.

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3. U.S. Department of Energy and Tennessee Valley Authority, “Interagency Agreement, DE-SA09-01 SR18976/TVA No. P-01 N8A-249655-001, between the Department of Energy (DOE) and the Tennessee Valley Authority (TVA) for the Off-Specification Fuel Project”, Modification 30, April 2009.