

DISPOSITION OF MULTIPLE FORMS OF HIGHLY ENRICHED URANIUM IN KAZAKHSTAN

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ABSTRACT

The disposition of highly enriched uranium (HEU) from various sources in Kazakhstan is an ongoing technical challenge. Since 1994, efforts have been made to find disposition pathways for HEU materials from various facilities in Kazakhstan, including the Ulba Metallurgical Plant, the Reactor in Aktau, the WWR-K Reactor at the Institute of Nuclear Physics (INP) in Alatau, the IVG.1M Reactor at the Baikal-1 facility near Kurchatov, and the IGR Reactor facility, also near Kurchatov. The Kazakhstan Ministry of Energy is responsible for the disposition of the materials, and through their operating entities they are developing disposition paths for the various materials. This effort is being conducted with the support of the International Atomic Energy Agency (IAEA), ROSATOM, and the U.S. Department of Energy (DOE) National Nuclear Security Administration's (NNSA) Office of Material Management and Minimization (M3).

The HEU is in many forms and therefore requires a variety of disposition pathways and approaches. In this paper we will discuss the various technical challenges associated with the disposition of a wide range of material forms and properties.

1 INTRODUCTION

Kazakhstan has a rich history of nuclear technology development, a program that includes fast, thermal, and fusion reactors; space rockets, isotope production, and materials research. Since the end of the Soviet Union, Kazakhstan has been working to eliminate many of the hazards associated with their legacy programs while preserving their viable capabilities. Kazakhstan entered into its most significant agreement¹ in 1993 with the objective of safeguarding and dispositioning all their nuclear weapons materials. The first efforts in-country focused on improvement of security at Kazakhstan nuclear sites. In the late 1990s, disposition of the large cache of irradiated plutonium-bearing fuel commenced in Aktau², resulting in long term storage. Subsequent disposition of fresh fuel from the Aktau reactor began in 2000, resulting in down-blending by 2005. In 2000 the IAEA initiated efforts to repatriate Russian origin research reactor HEU fuel, which resulted in the Russian Research Reactor Fuel Return (RRFR) program.³ This program supported the repatriation of HEU from the INP reactor, which was completed in 2017. Since that time, efforts have focused on the remaining materials at the Kurchatov facilities, an ongoing effort. In this paper we will discuss the various technical challenges associated with the disposition of a wide variety of material forms and properties since the year 2000.

2 Forms and Properties of the Materials

The nuclear materials in Kazakhstan include a range of fuel forms including power reactor fuel, plutonium production targets, research reactor fuel, and some unique forms. The fuels include conventional co-extruded uranium-aluminum fuel elements as well as zirconium-uranium alloys and uranium-impregnated graphite blocks. Irradiation ranges from high burnup to lightly irradiated to fresh material. Quantities also vary, from multi-shipment loads to small numbers of fuel elements. We will describe each portion of material and describe the disposition path developed, as well as the status of disposition efforts.

2.1 HEU Materials

Following completion of the down-blending of the Aktau fresh fuel in the late 1990's, it was recognized that other unirradiated HEU materials existed at the INP in Alatau, which would require down-blending. These materials were of different forms and quantities, however, and new capabilities were needed at the Ulba facility to deal with the multitude of forms. Ulba developed processes to deal with various forms of HEU materials, including UO₂ pellets, powders, UF₆ in sealed containers, aluminum/uranium solid fuels, and experimental fuel assemblies. In 2006 efforts were initiated to develop processes and equipment to facilitate the down-blending of these materials. By May 2011 the processes and equipment were ready for operation, and a total of 33kg of material was shipped to the Ulba facility. Processing of the materials was completed by the end of 2011, and the material was returned to the INP facility.

2.2 INP Fresh Nuclear Fuel

The largest portion of the initial 33kg of unirradiated HEU materials were regular and experimental fuel assemblies for the INP VVR-K reactor in Alatau. The form of the material was typical of VVR-K fuel assemblies, a billet of Uranium-Aluminum alloy co-extruded with aluminum to form the assemblies. Unlike the Aktau solid oxide pellet form, the solid aluminum/uranium fuel assemblies required a much more extensive process to recover the ²³⁵U in the fuel. The process developed at Ulba started with cropping of the fuel to remove excess aluminum portions of the fuel assembly. The next step was to dissolve the assembly in nitric acid. The product of this step was filtered to ensure complete dissolution, with remaining particles returned to the dissolver. Uranium was then precipitated from the resulting solution, dried, and oxidized to produce U₃O₈ powder. This powder was then mixed with unenriched U₃O₈ powder in sufficient quantities to lower the enrichment to the desired 19.8-19.92%. To ensure uniformity of materials, this combination was then again dissolved, filtered, and calcined to produce a uniform U₃O₈ powder of the correct enrichment. The finished product was then packaged in containers for storage and transport to INP.

Subsequent batches of unirradiated VVR-K fuel were shipped in 2014 (10.2kg) and 2016 (6.5kg) for down-blending. Down-blending of the last batch was completed in December 2015 - January 2016. Over a period of 5 years, the three batches of fuel assemblies totaled 49.7 kg.

2.3 IGR Spent Nuclear Fuel

Construction of the IGR reactor, a pulse-type test experimental reactor, was initiated in 1958. First criticality occurred in 1960, with full power reached in 1961. The reactor contains a center axial experimental hole that is subjected to a large (10GJ) pulse of neutrons when the reactor

is operated. The reactor is operated by quickly removing the control rods, resulting in a large pulse of neutrons which is terminated in a fraction of a second by the negative coefficient of reactivity. In 1967 the reactor was modified to increase the loaded mass of ^{235}U from 7.46 kg to 9.0 kg, as well as to increase the size of the main axial experimental center section from 180mm to 290mm.

The 7.46 kg core from the first period of operation is currently stored onsite and is the subject of the first disposition investigation. The fuel form for the IGR reactor is unique. It is fabricated by soaking graphite blocks with a solution of uranyl-dinitrate ($\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$), impregnating the graphite with 90% enrichment uranium. Processing this material is not straightforward, as the energy associated with consumption of the graphite (the nuclear ratio of $\text{C}/^{235}\text{U}$ is 8040) can prove very difficult in a reprocessing facility. In addition, the byproducts resulting from various approaches to separating the graphite from the uranium are problematic.

After the initial investigation of existing processing methods, four approaches were selected for further study⁴; chemical technologies, dry mixing, combustion, and combined combustion and chemical. The dry mixing proved to be the least problematic, as it did not involve adding energy to the carbon, requires much simpler equipment, and has no potential for separation of the HEU in the process. This approach has been selected for further development and for assessment of suitability for disposal.

Although the IGR fuel in question is irradiated, the fluence is lower than typical spent nuclear fuel, and for this reason an approach that utilizes gloveboxes to accomplish the down-blending process was chosen. Currently under development, the process will involve crushing the uranium-graphite blocks to powder-like form and mixing it with unenriched uranium powders to obtain an LEU product. This product will be mixed with an optimized combination of fly ash, slag, cement, and water to create a grout that can be mixed in drums. The final product will have sufficiently low uranium concentrations to dispose of without the need for continuing application of safeguards.

Transport will not be an issue, as the process will be implemented in-situ. Conversion of the IGR reactor to LEU is currently under consideration, and any HEU fuel discharged will likely be dispositioned in a similar manner as above.

2.4 IGR Fresh Fuel

Unirradiated uranium-graphite fuel remained from the modernization effort in 1989, with about 2.9 kg of material remaining on site. Based on the value of the ^{235}U contained, a decision was made to develop a down-blending process for these materials. The Ulba Metallurgical Plant was tasked with developing the down-blending process, which involves fuel crushing, oxidation of the graphite, oxidation of the uranium to U_3O_8 , and mixing with natural uranium U_3O_8 powder. This was completed in 2019, and the materials were shipped to Ulba for processing in late 2019. After about six months of processing, the down-blend was completed. The resulting ~19% enriched LEU powder was dissolved in nitric acid and the resulting uranyl nitrate solution will be used for further processing at the Ulba facility. It should be noted that the relatively small quantity of material made the oxidation approach feasible as compared to the cementation approach applied for the irradiated graphite fuel discussed in section 2.3.

Transport of the materials from the IGR facility to Ulba was straightforward. Standard fresh fuel packages were loaded onto trucks for the long drive to Ulba.

2.5 INP Spent Nuclear Fuel

The WWR-K reactor in Alatau near Almaty has operated since 1967, and although shipments of spent nuclear fuel had occurred in the past, by 2004, a substantial quantity of fuel assemblies had accumulated onsite at the INP facility. The INP WWR-K reactor facility is a typical Soviet design, which incorporated measures to allow for loading and transport of spent nuclear fuel for shipment to the Soviet Union. The fuel, uncropped, is loaded from the facility spent fuel pool into a basket holding four WWR-K assemblies, which is then loaded into a TUK-19 cask, purpose-built for transport of WWR-K fuels. The casks were then transported by truck to the railhead, where they were loaded in special railcars designed to carry the TUK-19. The rail journey of 1400 miles ends at the Mayak facility in the Russian Federation, where under the RRRFR program, the fuel is dissolved for recovery of the ^{235}U . Because the number of TUK-19 casks constructed is limited to 20, four shipments were required over a period of six months in 2008-2009 to accomplish removal of the first tranche of 278 spent fuel assemblies bearing 73.7 kg of HEU from INP.

In 2014, another portion of the fuel, now sufficiently cooled, was transported using the same disposition pathway. The mode of transport was altered to air shipment, however, to minimize the security risk during transport. For this effort, the same TUK-19 casks were used, but placed within specially designed 20-foot ISO containers to allow for ease of handling by the aircraft, the Antonov-124-100. Each ISO container held two or three TUK-19 shipping casks, and five ISO containers were used to ship the 16 casks in each shipment. Two air shipments were used to transport 127 assemblies containing 37.3 kg of HEU to Ekaterinburg, Russia, from which they were transferred to trucks for the final ground leg to the Mayak facility for storage and reprocessing for recovery of the uranium. One item of note for this shipment was that the 16 TUK-19 casks arrived at Mayak and within two weeks were back at INP, loaded, and ready for the second shipment.

Following successful demonstration of the use of LEU lead test assemblies in 2014, the remaining HEU fuel was discharged en-masse and the reactor was converted to LEU fuel. Following sufficient cooling time, the remaining HEU fuel was shipped to Mayak in a final round of three air shipments from July thru September 2017. These shipments utilized the same approach of loading two or three TUK-19 casks in an ISO container and then loading the ISO containers on the Antonov. Overall, 47.3 kg of HEU spent nuclear fuel was shipped to the Mayak facility for reprocessing in 2017, eliminating all HEU from the INP facility.

2.6 IVG.1M Reactor Spent Nuclear Fuel

The IVG.1M Reactor was developed as a nuclear rocket prototype, commencing operation in 1972 and reaching full power (72MW) startup in 1975. The reactor was originally hydrogen gas cooled but was later refitted with water cooling jackets in 1989. The fuel used for the IVG.1M reactor is a billet of uranium-zirconium alloy co-extruded with a Zr-Nb alloy cladding.

Efforts to remove the HEU spent fuel from the IVG.1M were initiated in 2014. A feasibility study was conducted to develop a process for dissolution of the uranium-zirconium alloy fuel at Mayak. The study identified modifications to the chopping and dissolution approaches that would allow the fuel to be reprocessed. Current efforts are in progress to modify the license and physical facilities at Mayak to accommodate the new processes. Once this is accomplished, and the facility is appropriately prepared, the fuel will be shipped.

Preparing the fuel for shipment at the IVG.1M reactor is much more involved than typical research reactors. The fuel rods consist of twisted extrusions which are bundled together and placed in a much larger carrier assembly that also serves as the coolant channel for the fuel. Shipment of the larger assembly is impractical and would require an extremely large (and therefore heavy) shipping cask. As a result, the fuel must be cropped prior to shipment.

The fuel assemblies are individually transferred from the reactor to a storage area adjacent to the hot cell facility. They are then individually transferred to the hot cell where the fuel is cropped top and bottom to segregate the fuel-bearing portions from the larger assembly. The fuel portion is then loaded into a canister, which is then loaded into the basket of the TUK-19 shipping cask. Once loaded with three canisters, the basket is then removed from the hot cell using a second transfer cask which is designed to mate with the TUK-19 Cask. This smaller transfer cask is moved from the hot cell to a separate room used for staging the TUK-19 casks. Once the basket is transferred to the TUK-19, the cask is sealed, dried, and leak tested. At this point the TUK-19 is transferred to an outside staging area where it is loaded into ISO containers. These ISO containers are then loaded onto a truck for transport to the railhead, where the ISO containers are loaded for shipment to the Mayak facility in the Russian Federation. At Mayak, the fuel will be stored until ready for processing, and then reprocessed to recover the ^{235}U for use in LEU fuel.

3 LESSONS LEARNED

The wide variety of fuel forms found in Kazakhstan has provided an unprecedented challenge for determining disposition pathways. The key lesson learned is that forward-looking assessments can greatly simplify and speed up the disposition process. Technical capabilities, once developed, should be given serious consideration for retention for future use even if no immediate need is on the horizon. Portions of the versatile Ulba facility were faced with an uncertain future but were not eliminated and were subsequently available for use in addressing the multitude of challenges eventually presented.

4 CONCLUSION

The wide variety of materials associated with the nuclear industry in Kazakhstan called for an equally wide variety of solutions. Combining tried-and-true approaches used in other countries with development of unique processes has allowed Kazakhstan to move forward with their disposition of nuclear materials in a timely and cost-effective manner. Consideration of a multitude of options, including both existing and conceptual approaches is essential to finding the best solution for disposition of nuclear materials.

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