Indonesian Highly Enriched Uranium-Bearing Waste Elimination - 17393

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ABSTRACT

The Department of Energy's National Nuclear Security Administration (NNSA) and the Y-12 National Security Complex (Y-12) have been working with PT Industri Nuklir Indonesia (PT Inuki), an Indonesian medical isotope producer, since 2014 to disposition all highly enriched uranium (HEU) stored at the government-owned Research Center for Science and Technology (PUSPIPTEK) near Serpong, Indonesia. To eliminate the 1.5 kilograms of liquid HEU at the facility, Y-12 proposed a method to down-blend the leftover process solutions with a depleted uranium solution made on-site to render low-enriched uranium (LEU). This method is essentially irreversible since the uranium enrichment is degraded using a molecular level of intimate mixing in aqueous solution.

This cooperation was tied to a presidential commitment made at the 2016 Nuclear Security Summit to down-blend all remaining HEU in Indonesia by September 2016. While all unirradiated HEU was down-blended to LEU in March 2016, the campaign to down-blend the 1.4 kilograms of irradiated HEU began in late July 2016 and was completed in August, making Indonesia and all of Southeast Asia free of HEU.

As part of this cooperation, Y-12 and NNSA also proposed a subsequent disposition option for the LEU solution whereby the irradiated and unirradiated solutions will be chemically manipulated to prepare a dry LEU oxide suitable for interim storage and yield a uranium-free liquid stream that can be discarded as radioactive waste. In the fall of 2016, Y-12 and NNSA plan to begin developing the LEU solution disposition methodology with the Indonesian facility using small-scale batch-wise methods within existing hot cells, involving straightforward precipitation, ion exchange, decantation, and calcination methods.

INTRODUCTION

For over twenty years, the U.S. Department of Energy's National Nuclear Security Administration (NNSA) has worked with foreign partners to reduce stocks of weapon-usable nuclear material around the world. Today, NNSA's Office of Material Management and Minimization (M3) presents an integrated approach to addressing the persistent threat posed by nuclear materials through a full cycle of materials management and minimization efforts. As a result, M3's Office of Nuclear Material Removal has removed or confirmed the disposition of over 7,000 kilograms of weapon-usable nuclear material from six continents.

The Nuclear Security Summit process has further motivated efforts to eliminate vulnerable weapon-usable nuclear material by convening heads of state to discuss progress and commit to increasing nuclear security globally [1]. At the 2016 Summit, President Obama and Indonesian Vice President Kalla announced the successful down-blending of all unirradiated HEU to low enriched uranium and committed to down-blending the remaining 1.4 kilograms of irradiated HEU by September of that year [2]. In August 2016, NNSA announced that Indonesia was the 30th country plus Taiwan to have eliminated all remaining stocks of HEU [3]. Significantly, this down-blending made all of Southeast Asia free of weapon-usable nuclear material.

To achieve this milestone, NNSA, with technical expertise provided by the Y-12 National Security Complex (Y-12), worked directly with the medical isotope production facility in possession of the material, PT Inuki, over a period of two years. PT Inuki ceased using HEU to produce the medical isotope Mo-99 several years prior but was left with hundreds of bottles of irradiated liquid HEU target residue material from its former production operations taking up space in its hot cells at PUSPIPTEK [4]. As it prepared to restart isotope production using LEU, the facility faced major space constraints for future radioactive waste streams. Therefore, NNSA proposed a cooperation whereby PT Inuki would eliminate its stocks of HEU in exchange for technical assistance addressing the residual liquid waste and developing a sustainable process to manage new waste streams [5].

DESCRIPTION

HEU Materials Description

Both the unirradiated and irradiated HEU solutions stored at PUSPIPTEK were a result of PT Inuki's Mo-99 production activities. The Mo-99 production process consists of electro-deposition of HEU as a thin metallic foil, irradiation of the foil in a research reactor, then dissolution of the foil and chemical separation to harvest Mo-99. The HEU by-products resulting from this process were comprised of two types of aqueous solutions: dilute unirradiated target plating solutions; and radioactive fissile waste (RFW) resulting from the processing of the irradiated HEU foils. The dilute unirradiated solutions were stored in multiple 20 Liter polypropylene jerry cans in an on-site laboratory. In total, these 11 jerry cans held approximately 110 grams of HEU dissolved in approximately 170L of solution. The irradiated RFW solution batches were stored in over 500 250mL glass bottles inside the processing hot cells at the radiochemical laboratory. Each bottle represented the waste from processing one foil and contained 2-6 grams of HEU in about 150mL of solution each.

Due to years of storage in a high radiation zone, nearly all of the paper labels that identified the RFW solution batches had deteriorated or decomposed. Additionally, the normally transparent polymer coating on the RFW bottles originally intended to prevent glass breakage had in most cases turned a dark opaque brown preventing visual observation of the bottle contents. Several RFW batches were split into two waste bottles. These factors presented several challenges during the irradiated down-blending campaign.

Processing Alternatives

In the early stages of this project, several options, including physical removal of the material from Indonesia, were considered. Based on the success of previous removal efforts, Y-12 initially planned to convert the HEU into a solid form that would be fit for shipment. This method involved using precipitation, filtration, and calcination to convert the HEU to an oxide and then remove the material to the United States for storage. While this process had proven successful on other projects involving HEU not containing fission and daughter products, it was determined that because of the irradiation history of the material, it would be neither feasible nor timely to decontaminate the HEU sufficiently to meet the U.S. storage facility acceptance requirements. Instead, a decision was made to eliminate the HEU by down-blending the U-235 content with either natural (NU) or depleted uranium (DU). The down-blending approach would accomplish the nonproliferation goal of converting the HEU to non-weapon-usable LEU, but defer the facility’s goal of removing the large volume of material from valuable work space at the radiochemical laboratory.

The down-blending was planned to occur *in-situ* by adding dissolved NU or DU (which only contain 0.7 and 0.2 percent of U-235, respectively) to the existing containers in measured amounts to drive the U-235 content to less than 20 percent—the enrichment limit for LEU. In early conversations with PT Inuki, suitable NU was identified at PUSPIPTEK but was held by another organization and attempts to transfer the material proved complicated. In later meetings, NNSA learned that PT Inuki held its own stocks of DU, which was eventually used to make up the source solutions. By using the official accountancy data of each batch, NNSA and PT Inuki planned to mathematically determine the amount of DU solution to dose each batch to bring the enrichment below 20 percent U-235.

Organizations Involved

Planning and execution of the proposed down-blending project required the agreement, cooperation and significant investment by several institutional participants from both the United States and the Republic of Indonesia. The U.S. team was led by NNSA M3 staff and relied on technical support from the Y-12 National Security Complex for definition of chemical, radiological, and nuclear material methods. PT Inuki led the field execution activities as the material and facility owner. PT Inuki was also responsible for transporting the DU from its storage facility to the laboratory and all related safeguards activities associated with material movement and modification. BATAN provided analytical services to verify the down-blending of the unirradiated solution. Regulatory oversight and licensing approval for the non-routine radiological operations were granted by the Nuclear Regulatory Agency of Indonesia (BAPETEN). Inter-governmental support for the project was provided by U.S. Embassy Jakarta and the Indonesian Ministry of Foreign Affairs (Kemlu). HEU transactions were monitored by the International Atomic Energy Agency (IAEA).

All of the coordinating agreements, documentation, field actions, and project schedule needs required constant communication and numerous visits to the facility to monitor progress and identify and resolve a variety of issues that arose during the project.

Processing Description

Down-blending is best accomplished in the solution state as a gas or liquid (aqueous solution or metallic melt). Solution blending ensures intimate comingling of the HEU and NU/DU at the molecular level and, therefore, precludes separation of the original materials by normal chemical or mechanical means. The regulatory target is less than 20 percent U-235, but a practical target of 18 percent was chosen to account for uncertainties introduced by measurement and laboratory practice.

The isotopic diluent selected was natural or depleted uranyl nitrate solution since the HEU was stored in aqueous solution form. The philosophy of the solution down-blending process was driven by a desire to minimize impact to the operation and utilize existing materials, skills, and equipment to the extent possible. To this end, the depleted uranium oxide (U_3O_8) that was available on site was chosen as the isotopic dilution source and an existing laboratory adjacent to the HEU storage area was the designated location to prepare the DU doping agent. Additional laboratory equipment was needed for this process, including beakers, hot plates, a scale, and several hundred appropriately-sized syringes for the hot cell application.

The unirradiated solution could be handled in the laboratory using minimal personal protective equipment (PPE) but the irradiated material stored in the hot cells required handling and manipulation with shielding provided by the hot cells to provide worker radiological protection. The hot cell processing added complications to both preparing for and performing the down-blending. For example, several remote manipulator arms in the hot cells that had not been used regularly in five years needed to be repaired before operations could begin.

Normal processing for Mo-99 separation involved the use of syringes to add reagents to the process containers, so it was decided to use syringes to add the DU solution directly to the RFW storage bottles as-is to simplify the process and to minimize the generation of additional radioactively contaminated container waste.

Compared with the simple process of pouring measured amounts of DU solution into the jerry cans in the laboratory, the process of adding the DU solution to the irradiated bottles was much more complicated. PT Inuki operators introduced pre-loaded syringes of DU solution into the hot cells through an entry cell in batches of 5 to 10 syringes at a time and used them to inject the DU dopant into each RFW bottle through the sealed rubber septum using the remote manipulators.

Because of the missing labels and bottle discoloration, the precise quantity of HEU and available volume in each bottle was not known but was limited to 100mL or less. This necessitated producing a DU solution that was very concentrated to ensure that sufficient isotopic dilution was accomplished without exceeding the bottle volume. Therefore, the dose for all bottles was determined by calculating the amount needed to bring the enrichment of the bottle with the greatest HEU content down to 18 percent and applying this maximum dosage to all of the RFW bottles. This method resulted in one bottle at 18 percent U-235 and the others well below that target.

DISCUSSION

Down-blending Process Discussion

To produce the DU solution, depleted uranium oxide was transported from another on-site storage area and dissolved in nitric acid on a hot plate in a fume hood. To down-blend the unirradiated HEU, the DU solution was prepared to a concentration of about 250 gU/L since there was sufficient available volume in the storage containers for the additional DU solution volume. For the smaller RFW bottles with limited available volume, the DU solution was prepared to a concentration of 400 gU/L.

The HEU content of the unirradiated solution batches was evaluated to readily determine the DU doping requirements; however, process history indicated that the multi-container batches might not be homogeneous. This required some mixing of the containers to ensure that the doping requirement could be safely divided equally among the multiple containers.

As mentioned previously, most of the batch identification labels on the RFW bottles had deteriorated or fallen off to the point that tailored batch doping was not possible. The accountancy data was studied and the doping quantity was pegged at the "worst case" (i.e., greatest quantity of HEU) batch loading. All of the bottles were doped with the same quantity of DU solution since the lack of labels made it impossible to otherwise guarantee that all of the bottles were down-blended to below the 20 percent U-235 threshold using an 18 percent target. Consequently, the DU solution requirement was 50mL at 400 gU/L. This resulted in a feasible combination of syringe availability, manipulator-friendly handling, and solution volumes to avoid transfer from the existing 250mL RFW bottles.

Schedule Issues

The unirradiated down-blending project schedule was motivated by the March 2016 Nuclear Security Summit, while the irradiated material down-blending schedule was defined by the agreement between the United States and the Republic of Indonesia during the Summit to down-blend all remaining HEU in Indonesia by September 2016.

PT Inuki had to meet several regulatory requirements before either the unirradiated or irradiated HEU could be down-blended. First, since the material was in part U.S.-origin HEU and the United States and the Republic of Indonesia have an Agreement for the Peaceful Uses of Nuclear Material (also known as a 123 Agreement), a subsequent arrangement to modify the material in form or content was required.

BAPETEN required that PT Inuki perform a significant renovation of the radiological exhaust system prior to granting the domestic license for fissile material handling to ensure appropriate worker and environmental protection. This portion of the project alone had several funding, procurement, testing, and commissioning issues that threatened the success of the project but were resolved through diligent attention. Other pre-operational requirements included upgrade of the local area radiation monitoring equipment and preparation of the license application describing the planned down-blending tasks, safety procedures, and mitigations.

Though some of these tasks were required for both the unirradiated and irradiated down-blending processes, in the final week before the Nuclear Security Summit, PT Inuki was able to complete sufficient upgrades to satisfy BAPETEN that it could safely down-blend the unirradiated material, permitting the completion of this task before the Summit. The remaining repairs to the ventilation system and additional license application took time that compressed the processing schedule for the irradiated HEU into the summer of 2016.

Since the facility had not produced Mo-99 since 2010, it faced funding limitations that necessitated financial assistance from the United States in order to achieve project completion on the agreed timeline. Funds were used to purchase equipment and support payment for personnel activities to ensure timely execution of the down-blending project. A scope of work was quickly prepared to cover activities such as training preparation and execution, laboratory equipment procurement, waste clean-up and removal from the hot cells, as well as the salaries for laboratory workers to execute the material handling tasks, including over-time.

Unirradiated solution down-blending was performed in a laboratory using a separate hood ventilation system that was not dependent on the building ventilation system but still required the addition of a HEPA filter to accommodate the use of the powdered depleted uranium U_3O_8. Solution preparation and down-blending of the 11 jerry cans were completed in two days immediately following BAPETEN’s issuance of the license. These activities were completed in late March 2016, in time to be announced by President Obama and Indonesian Vice President Kalla at the Nuclear Security Summit.

The irradiated RFW bottle down-blending had to await the completion and testing of the building ventilation system as well as the issuance of the second operating license by BAPETEN. The RFW down-blending work did not commence until late July 2016.

Logistics

The irradiated down-blending staff was divided into three teams that acted in parallel to complete the work as expediently as possible. One team prepared 14 batches of DU solution in an adjacent laboratory that were hand-loaded into the 50mL syringes. A second team performed the transport logistics, moving the loaded DU doping syringes into the hot cells via the entry cell, moving and rearranging the RFW bottles to the designated hot cell by conveyor, and removing used waste DU syringes. The third team performed the actual down-blending in the hot cells and recorded the progress in a log-book to document completion. The U.S. participants observed all of the tasks and offered assistance as needed to resolve technical questions or problems.

Several operational difficulties were encountered that presented obstacles to the down-blending but were overcome through teamwork and innovation. Given the large quantity of bottles to down-blend (over 500) and repetitive nature of the process, even seemingly small problems had the potential to become major challenges. The large 50mL syringes were more difficult to push the plungers using the remote manipulators than the normal 10 to 25 mL syringes the operators were accustomed to using for Mo-99 production, which added time to the down-blending process. This problem was largely mitigated by using larger bore syringe needles that eased solution flow and employing an additional needle to allow the bottle to vent while the solution was injected. Many of the bottles had rubber septa that had become hardened over the years of storage that would not permit the insertion of the larger gauge syringe needles or plugged them with rubber. This required the use of a long pin inserted into the bore of the needle to clear the needle or replacement of the rubber septum and aluminum retainer cap. Because all of these intricate tasks were performed with remote manipulators, such mitigating actions doubled or tripled the time needed to process each bottle. The logistics of moving the large number of bottles from the storage cells to the operating cell and back again also required multiple actions to occur at any given time and was facilitated by the cooperative spirit, teamwork, and skill of the operating staff.

Reconciliation of the final LEU inventory for the irradiated solution items was simplified by the uniform DU dopant addition approach. Each item in the inventory could be adjusted by the fixed quantity of DU added to each bottle determined by the “worst case” doping methodology mentioned earlier.

LEU and Waste Disposition Alternatives

With the successful down-blending of Indonesia’s HEU, NNSA and Y-12 have begun coordinating with PT Inuki to determine an appropriate path forward to remove the solution waste from the limited hot cell and laboratory operating spaces. In October 2016, NNSA and Y-12 proposed several options for consideration by the Indonesian stake-holders, ranging from a “do nothing” baseline approach to full purification and recovery of the uranium.

Specifically, the options proposed were:

- Do not address the liquid waste (presented as a baseline for comparison against the other options);
- Relocate the materials to an appropriate, but lower value storage space;
- Dilute and discard the material (dependent on regulatory permissibility);
- Solidify through precipitation - convert the uranium values to an impure (possibly still highly radioactive) solid for more compact storage, while discarding the aqueous phase as non-fissile radioactive waste; and
- Purify and solidify – similar approach to the solidification option but including fission product removal to permit potential reuse of the uranium values.

PT Inuki and other relevant Indonesian stakeholders are currently considering these options to determine a viable strategy that addresses the legacy material and sustainably manages new irradiated waste from future Mo-99 production activities.

Lessons Learned

Multi-organizational international projects like this require special attention to adaptation, flexibility, and persistence. The original project concept and proposed methodology may require adaptation to the available materials and equipment, as well as established production methods and facility capabilities. It also must meet local regulatory and business requirements. Including these stakeholders early on improves communication and agreement. Because even the best laid plans can change, it is important to remain flexible and willing to adjust the project approach as new issues are uncovered from initial fact-finding to eventual implementation. Finally, integrating the needs and requirements of the various players to meet the overall objective requires persistence and frequent face-to-face communication. This is especially important when bridging different languages and cultural norms.

CONCLUSIONS

The solution blending approach developed by Y-12 and NNSA effectively and irreversibly down-blended Indonesia's HEU to LEU, meeting the nonproliferation objective of the two governments. By adapting the process to the facility's established production methods and using standard laboratory equipment, this achievement was reached at an acceptable cost and with minimal impact on the facility. Despite the technically simple approach, the project faced many administrative, logistical, and operational obstacles that PT Inuki, NNSA, and Y-12 overcame cooperatively. The relationships and teamwork developed over the course of the project were a major asset in solving problems as they arose. While some of these challenges impacted the initial project schedule, the team successfully met the 2016 Nuclear Security Summit commitments on time. Additionally, it is noteworthy that the down-blending process did not increase the facility's original waste inventory footprint. By injecting the DU solution into the existing bottles of liquid HEU, the number of containers and items on inventory largely remained essentially unchanged. The final disposition of this material will depend upon the waste management pathway PT Inuki chooses to pursue in the coming months.

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