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## FISSILE MATERIAL MINIMIZATION THROUGH PACKAGING AND REMOVAL OF PLUTONIUM FUEL MATERIALS

K.A. Dunn<sup>1</sup>, G.T. Chandler<sup>1</sup>, D.R. Leduc<sup>1</sup>, J.S. Bellamy<sup>1</sup>, T.J. Aucott<sup>1</sup>, D.W. Vinson<sup>1</sup>, and N.C. Iyer<sup>1</sup>  
<sup>1</sup>*Savannah River National Laboratory, Aiken, SC, USA*

R. Matzkin-Bridger<sup>2</sup>  
<sup>2</sup>*U.S. Department of Energy, Tokyo, Japan*

S.L. Dickerson<sup>3</sup>  
<sup>3</sup>*U.S. Department of Energy – National Nuclear Security Administration, Washington D.C., USA*

K. Hioki<sup>4</sup> and T. Yamamura<sup>4</sup>  
<sup>4</sup>*Japan Atomic Energy Agency, Tokyo, Japan*

### ABSTRACT

The pace of fuel cycle research with plutonium materials worldwide has decreased significantly over the last two decades and a number of research reactors using plutonium-bearing fuels have been shut down, resulting in weapons-usable plutonium materials at civilian research facilities. The United States, along with its international partners, has highlighted the need for a global commitment to minimize weapons-usable separated plutonium inventories.

In 2016, the U.S. Department of Energy's National Nuclear Security Administration (DOE/NNSA) and the Japan Atomic Energy Agency (JAEA) worked collaboratively to remove plutonium from the Fast Critical Assembly (FCA) reactor in Tokai-mura, Japan, which was shut down in 2011. DOE/NNSA and JAEA developed and implemented plans to characterize the fuel plates and rods using In Situ Object Counting System (ISOCS), package the materials in 9975 Type B nuclear material packages, and transport the fuel to a receipt facility for ultimate disposition. This joint campaign culminated in the world's largest removal of weapons-usable plutonium and was recognized as a major nonproliferation accomplishment by the global community.

This paper will describe the characteristics of the plutonium material and the technical activities related to characterization, packaging, and transport of the plutonium inventory. Specifically, it will describe in detail the isotopic characterization process of the plutonium materials, development of the packaging process flow sheets, design and development of the necessary containers for packaging operations, procedure development, and training of personnel. In addition, this paper will highlight the 9975 shipping package certificate validation process with the relevant regulators and review the material accountability and control and safeguards protocols practiced in the plutonium packaging, storage, and transport activities.

### INTRODUCTION

Civilian separated plutonium inventories worldwide are on the rise and recycling and/or disposition of plutonium continues to be a challenge. Civilian separated plutonium falls into two categories:

1. Separated plutonium associated with research and shut-down facilities, and
2. Separated plutonium associated with commercial nuclear power.

Plutonium from shut-down research facilities is a small subset of the global plutonium inventory, however, it is considered highly vulnerable because it is portable and in forms that are particularly attractive. Hence elimination of this plutonium is a high priority.

For over twenty years, DOE/NNSA has worked with international partners to reduce weapons-usable nuclear materials from around the world. DOE/NNSA's Office of Material Management and Minimization (M3) provides an integrated approach to addressing the persistent risk posed by nuclear materials through a full cycle of material management and minimization efforts. Through these cooperative efforts, M3's Office of Nuclear Material Removal has removed or confirmed the disposition of over 7,100 kg of weapons-usable nuclear material from six continents.

As new and pressing challenges arise, M3 has adapted its material minimization efforts to meet the task. For this reason, the Office of Nuclear Material Removal created the Gap program to address materials not included within the scope of the U.S.-origin or Russian-origin return programs. The Gap program addresses elimination of highly attractive unirradiated HEU and separated plutonium as well as irradiated HEU outside the scope of the U.S.-origin return program. Gap materials may be down-blended in-country or removed to a third country or the United States for downblending and/or disposition. In 2010, the necessary environmental approvals for the removal and disposition of separated plutonium were obtained. Since then, the DOE/NNSA/M3 Office has partnered with six countries to eliminate plutonium inventories in ten different facilities. The approach to the plutonium packaging and transport takes into account safely handling, stabilizing, packaging, transporting, and dispositioning of the separated plutonium. This paper will address the recent removal of plutonium from the Fast Critical Assembly (FCA) reactor in Tokaimura, Japan.

## **APPROACH**

The approach to the plutonium characterization, stabilization, packaging and transport for the FCA materials was different than previous plutonium removal campaigns because the plutonium material is hermitically sealed in stainless steel cladding and is considered engineered material.

Figure 1 provides a high-level roadmap for the characterization, packaging and removal of nuclear materials through the Gap program. For the case of plutonium materials, initially the separated plutonium inventories are reviewed with the partner country to identify materials that may be eligible under the Gap program. The FCA materials consisted of engineered plates and rods. The content of the engineered material included metal or sintered oxide fuels which were considered to be stabilized and contained, as long as the cladding retained its integrity. Fabrication and vendor contractual documentation indicated that the plates and rods were fabricated with a high degree of Quality Assurance and to strict tolerances.

Excellent book values were available for the plates and rods. These book values were used as the basis for understanding the available inventory.

Additional considerations for acceptance during the initial assessment phase include the quantity, type, and characteristics of the material, the location, storage condition, attractiveness level, ability to meet receipt facility criteria and availability of disposition path. A subsequent assessment phase is then initiated where a detailed documentation of the plutonium materials characteristics is developed, the stabilization and packaging options are evaluated, and a detailed plan and high-

level schedule are generated. It is during this assessment phase that details of the host country capabilities are determined including facilities and personnel competency.

The FCA plates and rods packaging and transport campaign required some new equipment for characterization and packaging for the packaging process. Some key pieces of equipment included multiple weighing scales and the ISOCS equipment. JAEA used an existing hood along with appropriate personnel protection equipment during the packaging operations to ensure safety of personnel in case of a potential breach.

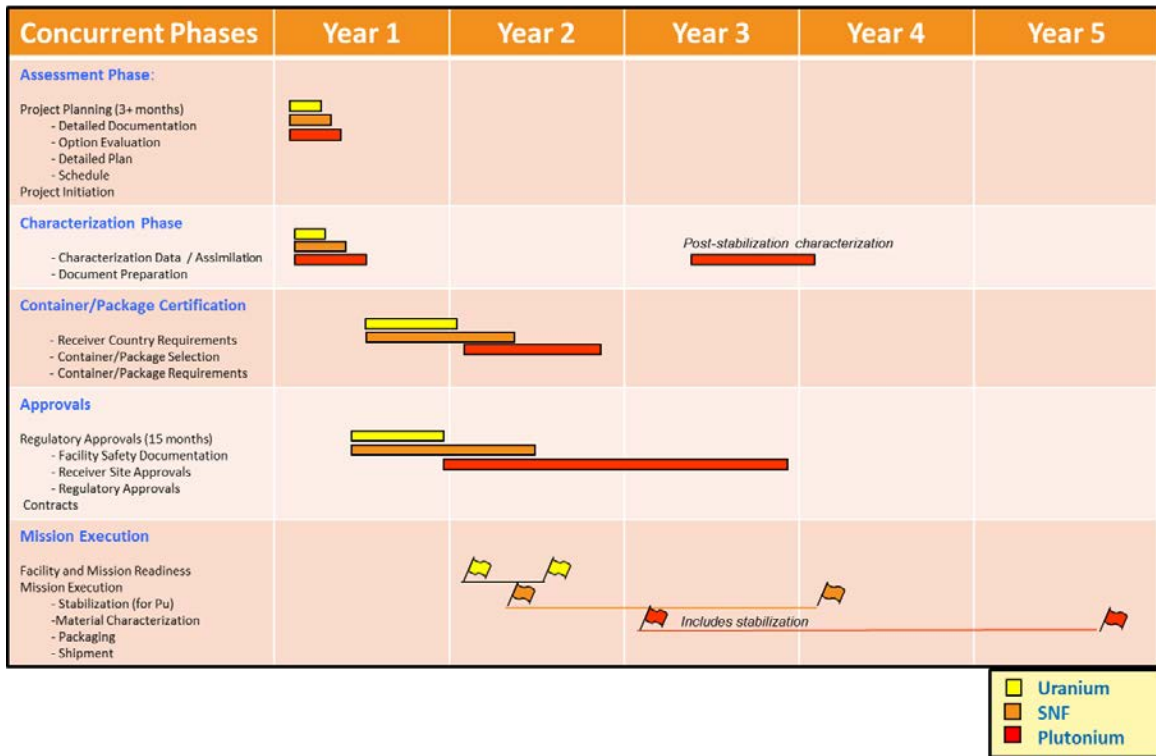


Figure 1. High Level Roadmap Example for Material Removal

Concurrent to the assessment phase, the host country is required to prepare a detailed description and storage history of the plutonium materials. Historical characterization data from the host country is also assembled. Additional characterization of the materials is performed in order to obtain as much information about the materials prior to their packaging and transport. A materials specific flowsheet is developed for the handling, characterization, processing, and packaging of plutonium material for each partner country. An example flowsheet for inspecting the FCA materials prior to packaging into the shipping packages is provided in Figure 2.

The regulators and key stakeholders at the packaging site are engaged throughout the planning process. Typically, throughout the project cycle, a number of regulatory approvals are required including facility safety basis documentation, receiver site documentation, and transport approvals. Figure 3 is a representation of the documentation required to be prepared by the packaging site (PS) in the partner country. There are three main sets of documentation that the packaging site must prepare that require a review and approval by the receiving site (RS). First,

prior to hot operations the packaging site develops the documentation that provides assurance that they comply with the receiving site acceptance criteria (AC).

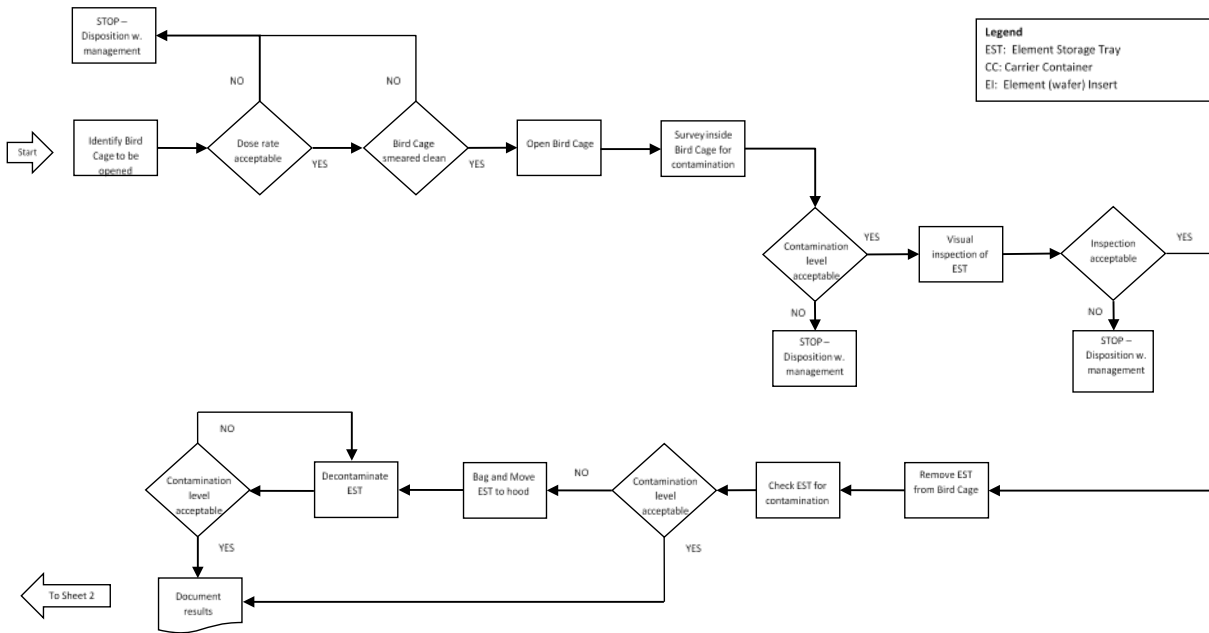


Figure 2. Flowsheet for Plate Transfer Process, Inspections, and Loading  
 NOTE: Inspections and weight checks looking for bulging, pressurization, weight gain, corrosion, intact welds, oxidation of metal.

Documentation Preparation	Preparation for Hot Ops		Hot Operations		Shipment of Materials
AC Documentation	—————				
PS Submit AC Documents to RS		♦			
RS provide letter for Hot Ops			♦		
PS Obtain Packaging Data			—————		
PS Submit Packaging Data to RS				♦	
RS Provide Letter for ATS				♦	
PS Assemble Package Documents					—————
PS Submit Remaining Package Documentation to RS – Project Complete					♦

PS – Packaging Site  
 RS – Receiver Site  
 AC – Acceptance Criteria  
 ATS – Authorization to Ship

Figure 3. Documentation that was Required from the JAEA FCA Reactor Facility

The packaging site is required to submit a documentation package to the receiving site demonstrating compliance to the receiving site Acceptance Criteria and obtain approval prior to commencement of the hot operations. The documentation package included a description of:

- FCA plutonium plates and rods to be packaged
- Compliance to ensure stabilization of the plates and rods
- Equipment being used for weighing and characterizing the plutonium materials including the acceptable performance of the equipment
- Containers and shipping packages to be used
- Procedures used for measuring and packaging the plates and rods
- QA program.

Key elements of the hot operations (i.e. operations which required handling of radioactive / nuclear materials) included material characterization and packaging. JAEA personnel at the FCA reactor facility collected data during the hot operations campaign that included materials form and characterization data including photographs, materials accountability data including measurement uncertainties, 9975 shipping package data sheets, dose measurements, etc.

During the packaging operations, JAEA performed air sampling on each bird cage to check for airborne contaminants prior to placement of the birdcage in the hood to inspect the fuel plates ensure the cladding retained its integrity. Once the bird cage was opened the inside was smeared to inspect for contamination. Clean limits were required prior to packaging. Each plate and rod was inspected visually for signs of degradation such as failed seam welds, oxidation of the plate or rod, and material build-up. Up to five (5) plates at a single time were loaded into the rack or holder. Once all of the plates and/or rods were assembled in the rack or holder for a particular container, the assembled rack or holder was smeared prior to placement into the carrier container to ensure no contamination was present. A thorough review of the fabrication specifications provided a high degree of confidence that appropriate QA was invoked at the time fabrication and the integrity of the plates and rods at the time of fabrication was verified. Additionally, prior to packaging of the plates and rods into the inner container of the 9975 package a visual examination as well as several contamination checks were performed to ensure no breach of the engineered material occurred. These steps ensured that the materials were stabilized because the plates and rods had maintained their integrity prior to shipment. The collected data was assembled by JAEA and provided to the receiver site for data validation and approval, resulting in an authorization to ship.

When the removal campaign was completed, all of the completed procedures and the raw data from FCA was compiled and sent to the Receiving Site for project completion.

## **MATERIALS CHARACTERIZATION**

### **Non-Destructive Assay Measurements**

As previously discussed, the book values for the plutonium was used the FCA plates and rods as the basis for characterization of the materials prior to packaging since they were hermetically sealed, engineered materials and their quality was deemed adequate. Additional characterization of the materials was done using an In-Situ Object Counting System (ISOCS), which is a non-

destructive gamma-ray measurement technique to validate the book values. The ISOCS measurement was done on the Gap materials container configuration prior to placement within the 9975 packages for shipment, Figure 4. This measurement is used to verify the total plutonium content and isotopic distribution of the material within the container, to ensure they meet the requirements of the 9975 SARP and the receiving facility requirements.

Since a detailed account of the FCA plates and rods was available with the book values a measurement strategy for each material sub group was developed to perform ISOCS at the packaging site on a subset of the FCA containers.

- For each sub group which consisted of 6 packaged cans or less, measure at 100%
- For each sub group which consists of greater than 6 packaged cans, measure a random 20% of the cans from the sub group

Each sub group was modelled in the ISOCS Geometry Composer software in order to determine the detection efficiency for every configuration. Cans were placed on a turntable at a distance of 1 meter from the ISOCS detector. 2 mm of cadmium was used as an attenuator in front of the detector. Data was acquired for 1 hour on each item, after which the plutonium-239 content was calculated using the Genie software. Two isotopic codes (FRAM and MGA) were used to calculate the plutonium isotopic content, as well as uranium-235 and uranium-239.

The measurement uncertainty of the ISOCS model was estimated to be 10% (at one standard deviation). The total plutonium content of each item, including isotopics, was verified to within two standard deviations.

At the receiving facility, each 9975 package was measured using a drum-sized neutron multiplicity counter (NMC). These measurements allowed for the receiving facility to verify the declared plutonium content, as well as to confirm that the nuclear material was shipped and received properly.



Figure 4. ISOCS Equipment and Measurement

## Mass Measurement

A very prescriptive weighing procedure was implemented during the packaging of plates and rods at the FCA reactor facility. Three scales were used for mass measurements at various stages during the packaging process and three sets of calibrated weights were used to ensure the proper accuracy of the scales. Each mass measurement was read by a primary checker and then verified by an additional person to ensure independency during the scale reading. Additionally, photos were taken of the scale readouts for every calibrated weight measurement and every mass measurement of the containers and material. This rigorous approach for the mass measurements provided a very robust process that minimized the probability that over filling the containers would occur and provided assurance that the plates and rods were packaged safely without a concern of an undetected overbatch.

## PACKAGING OPERATIONS

The plutonium materials were packaged in stainless steel screw lid containers (SLC) designed specifically for packaging of the fuel plates and rods. The height of the SLC ensured that only one container could fit within the primary containment vessel (PCV) of the 9975 shipping package. The plates were secured within the SLC in racks designed for the appropriate size of fuel plate. The rods were secured within the SLC in holders designed specifically for the MOX rods and the remaining 5 PuO<sub>2</sub> rods. This FCA container configuration is shown in Figure 5 and placement of plates is shown in Figure 6.

The JAEA utilized the robust Type B 9975 Shipping Package for transport, Figure 7 which was selected for a variety of reasons. The 9975 shipping package is a robust Type B package specifically designed for the transport of plutonium in both metal and oxide form. It is designed and built to DOE, DOT, and IAEA standards and requirements. It is approved for U.S. domestic shipments of Pu metal and oxide and has been demonstrated with the land transport of >6000 packages in the United States. It was also approved for Gap materials shipments from several countries.



Figure 5. FCA Carrier Can Configuration for Plates

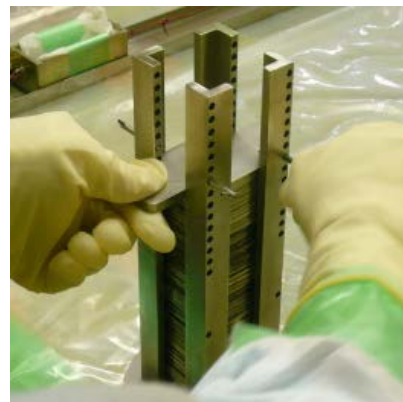


Figure 6. Placement of Plates in Rack



## 9975 PACKAGE VALIDATION

The DOE/NNSA/ M3 team worked closely with JAEA as well as Japan's Nuclear Regulation Authority (NRA) to ensure all appropriate reviews of the 9975 Safety Analysis Report for the Package (SARP) [Ref. 1] were completed and the U.S. certificate validated. In Japan, the NRA requires a separate submittal of the shipping package SARP for each shipment. This required the detailed review and validation of every analysis and test in the 9975 Safety analysis report that was related to the plutonium clad metal plates and clad mixed oxide rod materials shipped. The team responded to formal questions from both the JAEA applicant as well as directly from the Nuclear Regulation Authority in Japan.

The bounding plutonium contents in the 9975 packages presented several challenges. The bounding plutonium metal configuration in the 9975 has a calculated maximum  $K_{\text{eff}}$  effective with uncertainty ( $K_{\text{eff}} + 2\sigma$ ) of 0.945 which is one of the highest reactivity values found in transportation packages. The bounding plutonium oxide content assumes maximum radiolysis of moisture resulting in a maximum normal operating pressure of more than 300 psi. Although the actual plutonium metal plates and clad oxide rods would not result in approaching the conditions of the bounding plutonium contents, it was desired for the shipment to take place under the existing certificate and SARP. Supplemental analyses were performed which demonstrated that the worst case loading of FCA metal plates would result in a  $K_{\text{eff}}$  effective with uncertainty ( $K_{\text{eff}} + 2\sigma$ ) of less than 0.7 in all cases. To address worst case pressures and temperatures, another supplemental analysis was performed which included superposition of insolation with the fire heat load as required by regulations in Japan. Supplemental analyses were submitted in response to specific questions from either the JAEA or the NRA.

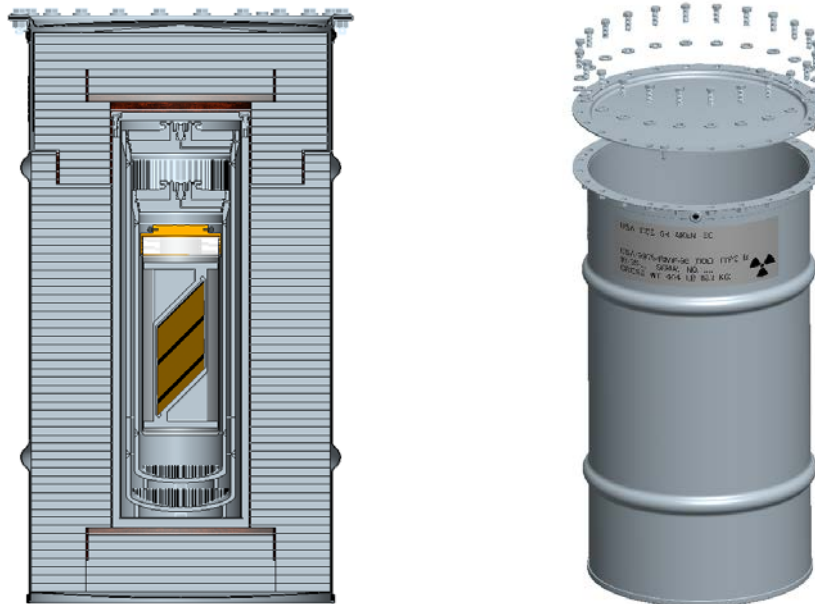


Figure 7. Type B 9975 Shipping Package

The 9975 package is a design iteration of a family of drum packages designated as the 99xx series that included eight variants. Because the certification testing was performed for the family as a whole, some tests were performed on other package variants and applied to the 9975 by

comparison. Because of the hazardous nature of the contents being shipped and differences amongst the tested packages, the NRA requested that a supplemental drop test be performed in the orientation that was most challenging. At the request of the NRA, SRNL performed the drop test requested of a 9975 package on June 2<sup>nd</sup> of 2015. The drop test validated that the 9975 is a very robust shipping package. Both the primary and secondary vessels were leak tested after the drop and found to be leak-tight.

## **TRANSPORT STRATEGY**

The transport strategy was developed to ensure the appropriate safety and security regulations were observed and consistent with IAEA standards for a category 1 transport. Calibrated equipment needed for packaging and sealing the FCA materials within the 9975 shipping packages was sent to JAEA. This approach ensured that JAEA packaged the materials in compliance with the receiver site requirements and per the 9975 SARP. Figure 8 shows the leak tester, a portion of the equipment, shipped to the partner country for validating that the containment vessels are sealed properly.

The FCA materials were packaged in containers which were placed within the 9975 shipping packages. The 9975 shipping packages were then placed within a cargo restraint transporter (CRT), Figure 9. A total of five 9975 shipping packages were placed within a single CRT. The DOE/NNSA/M3 Team provided assistance with development of the procedures and training to ensure efficient and effective loading operations.

The CRT pallets were then tied down with dedicated slings and fastened to special anchorage systems within ISO containers. Fastening of the CRT pallets to the ISO containers was designed to meet not only land transport requirements but also very strict maritime transport regulations for a Category 1 shipment, Figure 10.

In all packaging and transport cases the work was executed in compliance with the highest safety and security standards and regulations. Preparation for the physical protection and emergency plans was lengthy and complex. However, the partnership between the DOE/NNSA/M3 team and Japan's team ensured the transport of the materials was safely completed with an enhanced degree of safety and security measures, as required for a Category 1 Transport.

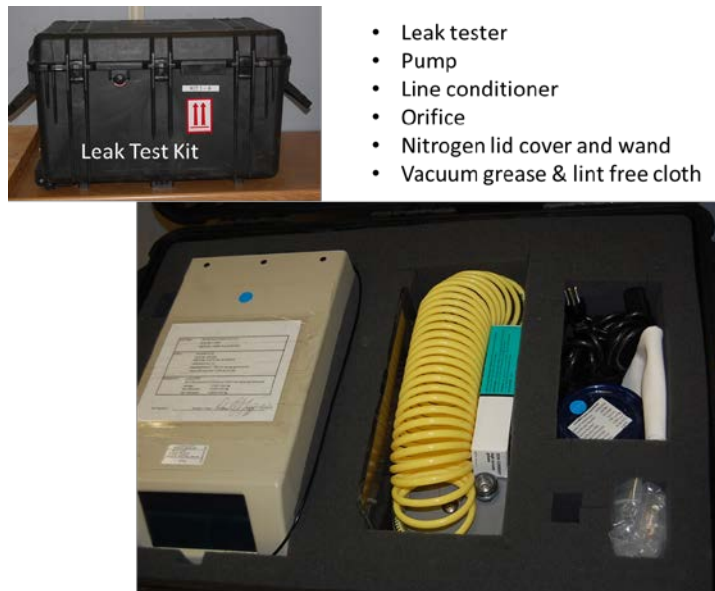


Figure 8. Leak Test Kit Loaned to Partner Countries

## CONCLUSIONS

Recent Nuclear Security Summits have highlighted the need for a global commitment to minimize weapons usable separated plutonium inventories. The United States worked with Japan and the JAEA to facilitate packaging and transport of plutonium materials from the FCA research reactor facility in Tokai-mura for eventual disposition. Additionally, since the quantities of separated plutonium predicated the use of Category 1 transport for these materials, tri-lateral cooperation was needed including international regulatory and security authorities to facilitate protocols for secure maritime transport of the plutonium materials. This plutonium minimization campaign from Japan was announced at the 2016 Nuclear Security Summits and was widely recognized as a significant nuclear security accomplishment.



Figure 9. Cargo Restraint Transporter



Figure 10. Land Transport with ISO Containers and Typical Maritime Transport Vessel

## REFERENCES

[1] Safety Analysis Report for Packaging Model 9975, S-SARP-G-00003, Revision 4, December 2015

## ACKNOWLEDGMENTS

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