

RADIOACTIVE WASTE MATERIAL CHARACTERIZATION BY NUMERICAL AND ANALYTICAL METHODS IN ANL-W RADIOLOGICAL AND NUCLEAR FACILITIES

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ABSTRACT

Argonne National Laboratory-West (ANL-W) is a research and development laboratory operated by the University of Chicago under the direction of DOE-CH. Primary research activities at ANL-W include nuclear reactor fuel and materials development, post-irradiation examination, fuel cycle research, spent fuel treatment, and assembly of space craft heat and power sources in support of NASA missions. Operating facilities at ANL-W include industrial, radiological, and category 2 and 3 nuclear facilities. Radioactive material is routinely shipped between ANL-W facilities as needed in support of research programs. This paper describes methods used to manage radioactive material transfers in a manner that prevents facilities from inadvertently exceeding radioactive material inventory limits allowed under their nuclear safety documentation safety basis.

The primary responsibility for nuclear safety compliance lies with each facility management team. A method was sought which would have as little impact as possible on daily facility operations, yet give facility management assurance that they were operating within prescribed limits. It was recognized that many transfers would involve such small quantities of radioactive materials that they would be insignificant to the nuclear safety of that facility. These shipments should require minimal effort to quantify and document the nuclear safety consideration. It was also desirable to use existing and common measurement techniques to quantify radioactive material transfers to the extent possible.

ANL-W uses various methods to quantify radioactive material shipments such as analytical lab sampling analysis and direct radiation readings. Analytical methods are straight forward but can be costly for all shipments especially routine facility waste transfers that are slightly contaminated. ANL-W has proceduralized a process using known waste stream isotopic composition coupled with shielding analysis to develop a correlation between radioactive dose rate and isotope curie inventory for specific package geometries and densities. Dose rate readings are already required for radioactive material transfers; this provided a good method to exempt small quantity shipments from full nuclear safety evaluations without adding a significant burden to shipping those packages. Shipments containing isotopes other than gamma emitting isotopes or which exceed a pre-determined radiation reading require further evaluation before shipment. Those shipments may be evaluated using radiochemistry analysis or through non-destructive assay techniques, both methods are available at ANL-W.

INTRODUCTION

Argonne National Laboratory - West (ANL-W) is a nuclear research and development laboratory operated for the Department of Energy (DOE) under contract by the University of Chicago for the past 55 years. Areas of research focus on national concerns such as nuclear energy development, nuclear safety, spent nuclear fuel treatment, non-proliferation issues, nuclear facility decommissioning and decontamination technologies, nuclear fuels development, and spacecraft heat and power sources. These research activities are conducted in a number of facilities at the ANL-W site including non-nuclear, nuclear category 3, and category 2 facilities.

Care must be taken to prevent non-nuclear and nuclear category 3 facilities from exceeding radiological material inventories appropriate for their nuclear safety classification as specified in DOE STD-1027-92. Of particular concern are shipments originating in nuclear category 2 facilities and being sent to nuclear category 3 facilities; or shipments originating in category 3 facilities and being sent to radiological facilities. A container of little concern in a given facility might contain enough radioactive material that when shipped to a facility with lower nuclear safety classification could cause that facility to exceed threshold values for the next higher nuclear safety classification.

With these concerns in mind ANL-W sought practical methods for tracking inventories and radioactive material transfers in each category 3 and non-nuclear facility. While the importance of keeping good inventories was clear it was not necessary to create undue burden on facilities by adding large amounts of record keeping or exhaustive analysis on shipments so small as to be inconsequential to any facility inventory in terms of nuclear safety. To this end, ways were sought to categorically exclude insignificant transfers from detailed record keeping. In order to do this however, affected facilities wishing to take advantage of this categorical exclusion would be required to maintain a "buffer" in their inventory that would allow storage of a large number of these excluded shipments without causing that facility to exceed its' nuclear facility classification.

Affected Facilities

DOE STD-1027-92 was issued to help DOE facilities complex wide consistently comply with the requirements of DOE Order 5480.23 *Nuclear Safety Analysis Reports*. DOE STD-1027-92 classifies nuclear facilities based on the facility's radiological and/or hazardous material inventory and the potential for off-site, on-site, or localized consequences. The following table details the categories of nuclear facilities specified in DOE STD-1027.

Table I. Categories of Nuclear Facilities Specified in DOE STD-1027.

Hazard Category	Description
Hazard Category 1 (HC-1)	Shows the potential for significant off-site consequences.
Hazard Category 2 (HC-2)	Shows the potential for significant on-site consequences.
Hazard Category 3 (HC-3)	Shows the potential for significant, but localized consequences.

There are no HC-1 nuclear facilities at ANL-W. HC-2 nuclear facilities are exempt from consideration for nuclear material inventories or transfers at this level since the unreviewed-safety-question process is sufficient to ensure that new programs are adequately reviewed and HC-2 facilities are maintained within the nuclear safety authorization basis for operation. Facilities which fall below the HC-3 threshold values are exempt from DOE Order 5480.23 but should still have controls in place to prevent HC-3 values from being exceeded¹. Therefore, the methods discussed in this paper are applied to non-nuclear facilities where radioactive material is stored or handled (referred to as “radiological” facilities) and HC-3 facilities at ANL-W.

Facility Inventories

Overall responsibility for material inventory at ANL-W facilities lies with the facility management. However, to assist the facility manager with this responsibility Radioactive Material Acceptance Coordinators (RMAC) have been assigned. RMACs and facility managers for each HC-3 and radiological facility maintain a database of facility inventory of each radionuclide found in that facility. This inventory is used as a baseline to determine the Sum of Fractions as required by DOE STD-1027. Sum of Fractions is determined as follows:

- for an HC-3 facility:

Sum of Fractions = [inventory of isotope A/HC-2 threshold of isotope A]+[inventory of isotope B/HC-2 threshold of isotope B]+[inventory of isotope n/HC-2 threshold of isotope n]

- for a radiological facility:

Sum of Fractions = [inventory of isotope A/HC-3 threshold of isotope A]+[inventory of isotope B/HC-3 threshold of isotope B]+[inventory of isotope n/HC-3 threshold of isotope n]

HC-3 threshold quantities of most radioactive isotopes are found in Los Alamos National Lab fact sheet LA-12981-MS². HC-2 threshold quantities of most radioactive isotopes are found in Los Alamos National Lab fact sheet LA-12846-MS³. These values are based on unmitigated releases of each individual isotope to a receptor at a downwind location and a specified committed effective dose (CED) uptake at that location.

Prior to any shipment of radioactive materials, the shipping facility is required to characterize and quantify the material being shipped. The receiving facility RMAC reviews and approves the characterization process. The proposed shipment inventory is then added to the known facility inventory and the Sum of Fractions is again obtained. If the resulting Sum of Fractions does not exceed 1, the shipment may be approved by the facility manager and the RMAC. The shipment is completed and the new material is added to the facility inventory.

DOE STD-1027 allows for the exemption of the following items/materials from the facility inventory:

- Sealed radioactive sources that are engineered to pass the special-form testing specified by the Department of Transportation in 49CFR173.469 *Tests for Special Form Class 7 Materials*, or testing specified in ANSIN43.6, *Sealed Radioactive Sources, Categorization*. (Documentation stating exempting standard must be retained by the facility in possession of such sources.)
- Radioactive materials used in exempted, commercially-available products as identified in 10CFR30.11 Specific Exemptions, through 30.19; such as timepieces, illumination devices, thermostats, electron tubes, microwave receiver tubes, etc.
- Materials contained in a DOT Type-B shipping container if the Certificate of Compliance is current and the materials stored in that container are authorized by the Certificate.

Characterization

Before any package of radioactive material may be proposed for shipment to another facility, sufficient characterization must be completed to provide the receiving facility enough information to make an evaluation for nuclear safety compliance. The proposed shipment must be characterized to identify each radioactive isotope and to quantify each of those isotopes. This characterization may be completed either by sample analyses, or by applying appropriate process knowledge, or a combination of the two methods.

Most of the radioactive material at ANL-W is generated as a result of either hot fuel examination, or reactor fuel production/development. Radioactive material generated from hot fuel examination typically consists of long lived activation and fission products from the EBR-II reactor fuel and is considered to be rather consistent in isotope concentrations. A two year hot cell smear survey campaign was used as the basis for determining isotopes and relative concentrations for this study. Samples from each zone in the hot cell were collected on a monthly basis and analyzed using multiple radiochemistry methods in order to measure gamma, beta, and alpha emitting isotopes. In this case, an extensive sample campaign is used to document acceptable process knowledge to be used in radioactive material characterization for hot fuel examination facilities. Undetected isotopes known to exist in equilibrium with other detectable isotopes were included in the pre-determined source term for this project. Using this process knowledge permits RMACs and facility managers to characterize radioactive material without re-sampling for each shipment.

Radioactive material generated from fuel development work generally uses isotopes of elements for which rigorous material accountability for security and safeguard purposes is maintained. For these processes mass balance records are maintained to account for the disposition of all materials. Containers generated under these programs are also examined using Non-Destructive Assay (NDA) methods which confirm the mass balance records noted above. These programs are often experimental in nature so the material composition assay is well documented. Using a combination of mass balance, NDA analysis, and material assay data results in good process knowledge, supported by analyses, sufficient to meet the characterization requirements prior to shipping radioactive material.

Facility managers and RMACs should be cognizant of operations conducted such that if new

programs introduce new isotopes or other process conditions change, these changes can be incorporated into the characterization process.

De Minimis Shipments

It was recognized early on in developing this process that there would be many very low level or “suspect” radioactive shipments that would have an inconsequential effect on the nuclear safety status of a facility. The impact to facility operations would be significant if these shipments were all held to the same level of evaluation and documentation as with other shipments containing larger quantities of radioactive material. A method of exempting the inconsequential shipments was developed reducing the unnecessary burden to facilities. It was highly recommended to use an approach to show compliance to DOE STD-1027 that could be accomplished in the field using existing methods and practices. It was decided that if portable health physics instrument radiation readings could be equated to a certain radionuclide inventory, it could be shown that any given package (meeting appropriate criteria) was below a pre-determined de-minimis inventory. An administrative level of 1×10^{-4} of the HC-3 threshold quantity was used as a de-minimis value - below which individual packages could be moved into HC-3 and radiological facilities without concern for compromising the nuclear safety of that facility. Criteria for meeting this de-minimis standard were established in order to “bound” which shipments could qualify for this exemption. These criteria include:

- Package contents are not shielded in any way except by self shielding of the package contents and the container walls.
- The radioactive material is known to consist of only mixed fission products with generation ratios similar to that generated in the EBR-II reactor.
- The package does not contain isotopes that are predominately alpha or beta emitters (e.g. uranium isotopes, transuranic elements, or pure beta emitters without a gamma tag).
- The package does not contain high density material such as soil, concrete rubble, or solid objects that would significantly shield the source term within the package.
- Packages considered for de-minimis could be only those common packages used to package, store, and transfer radioactive material. (Specific dimensions of poly bags, steel drums, poly sample containers, and wooden waste boxes.)

Limiting to those criteria, and using a commercially available radiation shielding model, a ratio of radionuclides was used which had previously been documented as a “typical” mixture of fission products found in ANL-W hot cells and in concentrations that would not exceed 1×10^{-4} of the HC-3 threshold quantity, a de-minimis radiation reading was determined. Using maximum allowable container weights for each container type allowed us to establish a maximum density for each container being considered for de-minimis shipments. Models were developed based on the above listed criteria and radiation readings were established for each package at the de-minimis value of 1×10^{-4} of the HC-3 threshold quantity.

It was found that the models (and actual readings) were very sensitive to the geometry of both the source and the package. If the source term were all placed in a small “hot spot” as opposed to being evenly dispersed throughout the contents of the container, the results could vary considerably. Or if the container geometry changed, such as by pressing on the sides of a poly

bag as opposed to merely measuring a reading at contact with the container's external surface would also result in differences in readings with the same source term. It was noted however, that 1 meter readings showed very little sensitivity to those same geometry changes. Accordingly, the preferred standard for using radiation readings to determine de-minimis source term packages was by using the 1 meter reading with a calibrated portable instrument.

Packaging commonly used at ANL-W for radioactive material storage or transfer include 1 liter poly bottles, four sizes of poly bags common to the nuclear industry, 55 gallon steel drums, and two sizes of fabricated wooden boxes. Models were developed for each of the common packages commonly used assuming that the pre-determined source term was evenly distributed throughout the volume of the container. The only shielding considered for this evaluation was self shielding of the source material and an insignificant amount of shielding provided by the containers' wall. Density for each modeled loaded container was based on maximum measured mass normally found for each of the respective loaded containers at ANL-W. Additional modeling was completed to measure the effect of placing the entire source term in a single location at various locations within the different containers, such as would be found with a radiological "hot spot". It was found that even though dose rates measured close to the container could vary significantly, the effect of hot spots on radiation dose rate was minimal for measurements taken at 1 meter from the container. The results of all cases modeled were within the detection range of portable instrumentation used at ANL-W making this a viable way to correlate dose rate to source term.

Once the modeling was complete, the process was peer reviewed and the modeled results were compared against other available shielding models. This independent verification indicated that the evaluation was correct in both principle and execution. The applicable concepts of this process were incorporated into a procedure to be used for all radioactive material transfers into ANL-W non-nuclear radiological facilities and nuclear category 3 facilities.

The transfers of de-minimis packages are allowed to be made with reduced characterization and documentation. For other low-level shipments not meeting the above exemption criteria, facilities may, on a case-by-case basis, develop methods to categorically exclude other similar low-level shipments. In these cases, documentation must show the methods used to make this determination.

CONCLUSION

ANL-W has successfully established a method whereby radioactive material transfers are documented and facility nuclear safety hazard categories are not exceeded. Considerable effort was expended up front to reduce the characterization and documentation burden for a majority of subsequent shipments which contain such small quantities of radioactive material as to be insignificant to the nuclear safety status of hazard category 3 nuclear and non-nuclear radiological facilities. By employing existing health physics practices and using shielding models, a de-minimis source term was correlated to a radiation dose rate at 1 meter for common packages. Packages with dose rates below the de-minimis values are managed with a lower level of characterization and documentation. Using this method has the advantage of continuing nuclear safety protection for the facility and allows resources to be devoted to higher activity

containers.

REFERENCES

1. DOE Standard 1027-92, December 1992
2. LA-12981-MS, Hazard Category 3 Threshold Quantities for the ICRP-30 List of 757 Radionuclides, August 1995
3. LA-12846-MS, *Specific Activities and DOE-STD-1027-92 Hazard Category 2 Thresholds*, November 1994