# REPACKAGING PLUTONIUM-238 HIGH NEUTRON DOSE RATE MATERIAL FOR WASTE DISPOSAL

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## ABSTRACT

Los Alamos National Laboratory reduced the Department of Energy's legacy nuclear material liability by repackaging problematic high-dose-rate plutonium-238 (Pu-238)/beryllium neutron emitters for deep geologic disposal at the Waste Isolation Pilot Plant (WIPP). The material presented two major hazards during processing – high dose rate and contamination. Undisturbed, the emitters' neutron dose rate was about 0.5 mSv per hour at contact. However, if disturbed as in crushing for repackaging, the emitters could produce a dangerous field of up to 50 mSv per hour at contact. Phase I of the project revealed that the material could be handled and packaged with an individual exposure of about 0.5 mSv per item or less whether the items were chemically dissolved or crushed and sieved. Phase II made nineteen items safe by removing the emitters from their original packaging and then crushing and sieving them into metal pipes that were placed into Pipe Overpack Containers for WIPP disposal. Waste disposal was chosen over reclaiming the Pu-238 because there is a simple path forward for the items as waste whereas there is currently no capability for purifying and reclaiming the Pu-238. The maximally exposed individual received less than a 0.07 mSv radiation dose for each item packaged. The characteristically high potential for contamination during handling Pu-238 was mitigated by the use of glovebags inside the glovebox.

## INTRODUCTION

The Department of Energy (DOE) has a large number of neutron emitters that have ended their useful life cycle and now must be disposed. These items do not present an imminent danger to humans in their present storage configuration, but their liability should be safely reduced in as timely and efficient a manner as possible.

When their service life has ended the emitters must be isolated from human contact or otherwise managed to reduce their hazardous properties. A project to dispose these items was conceived in two phases. Phase I was to characterize the material and determine how best to handle it, finding paths for all the waste streams, and securing permission to discard this material as waste. Phase II was to dispose of ten of the items, an amount that seemed reasonable given the programmatic budget.

The material could present two major hazards during packaging for safe disposal – high neutron dose rate and alpha contamination.

The neutron emitters are designed to produce a neutron flux by the interaction of alpha particles and beryllium metal. The neutron emitters repackaged in this project comprise about 4 grams of Pu-238 oxide micro spheres and 20 grams of beryllium metal micro spheres, either of which is coated with nickel. All the granules are immobilized in a matrix of potassium silicate or sodium silicate (water glass). In storage the items have an estimated dose rate of 0.7 mSv/hr. But the emitters are not sealed sources that would have a well-defined neutron flux. Instead they have the potential for producing up to 6.8 x 107 neutrons per second (or 50 mSv per hour at contact) when ideally mixed. Some mixing could occur during handling or shipping that could cause unacceptable radiation exposures to the workers.

It is widely accepted that Pu-238 oxide is highly dispersible and that strong control measures are required when handling it. Consequently, special facilities and methods were sought to avoid a release of material.

## ITEM DISPOSITION OPTIONS

Two disposal options were considered. The first was to dissolve the water glass matrix, separate the Pu-238 oxide for recycling, and dispose of the nickel, beryllium and water glass solutions as waste. The second option was to physically break up the mass to fit a new container and manage the entire package as waste for disposal.

The main advantage of the chemical dissolution method is that separating the beryllium from plutonium reduces the neutron dose rate potential to only that produced by Pu-238 oxide ($2 \times 10^5$ $sec^{-1}$), a desirable improvement in exposure safety. The increased risks of this method are 1) multiple handling steps, 2) management of not only dry waste streams but aqueous ones as well, leading to increased potential for an airborne release of plutonium, and 3) there is currently no capability for purifying and reclaiming the Pu-238.

The main advantage of the physical break up method is simplicity: fewer processing steps, less time spent fine-tuning the method, fewer waste streams to manage, and less likely exposure to contamination than with the dissolution method. Also, because all the waste is TRU waste, there is one path forward to WIPP for all the waste with no need for other disposal paths for non-TRU or aqueous waste streams. The main increased risk associated with the physical breakup method over the dissolution method is the unknown and probable increase in neutron (and gamma) dose rate during processing.

Two additional problems for the project were to secure permission to dispose of this material at all, and to dispose of attractiveness category D material[a] without additional processing.

## PHASE I: CHARACTERIZATION AND PLANNING

The purpose of Phase I was to retrieve an item from vault storage, examine the packaging, determine the handling characteristics of the material, and split the item into two fractions. There would be a sample fraction for further study and a storage fraction to be returned to the vault. The sample fraction would be dissolved, separated, analyzed, and the various waste steams managed. Information derived from Phase I would be used to make decisions about how to work off the ten items in Phase II.

### Ionizing Radiation Issues

The average measured neutron dose rate from the emitters in storage was about 0.5 mSv/hour at contact but the potential maximum neutron flux is 100 times that, 50 mSv/hr at contact (or 6.8 X $10^7$ neutrons per second), and would be a reason to handle this material in a hot cell rather than a glovebox. Whether the maximum dose rate, or even an elevated dose rate, would occur with this material was unknown. Would mobilizing the micro spheres by either crushing or dissolution of the matrix lead to increased dose? If so, how much? Would aqueous processing inhibit the alpha-n reaction and render the process eminently safe? How much dose would be acceptable? These were some of the questions answered in Phase I.

### Chemical Dissolution and Pu-238 Oxide Separation

In 2002 a single item was taken to a glovebox, unpackaged, and split into a sample fraction for testing and a larger fraction for immediate return to storage. The split was made to lessen the actual and potential dose rates during processing. It also reduced the amount of material at risk (MAR) in the laboratory so that neighboring operations could continue to work without exceeding the MAR limit of the building authorization basis.

In a glovebox, workers dissolved the sample matrix in an open beaker of heated 5 molar NaOH for about an hour, more if needed. They then size separated the micro spheres by washing the solids through two standard sieves – the plutonium was caught in one sieve and the beryllium in the other. They neutralized the rinsate and discarded it to a drain. Then they dried the beryllium spheres and disposed of them as solid transuranic (TRU) waste. The separated beryllium was sent to waste and the plutonium was consumed in subsequent analysis.

The analysis was a one-time occurrence and was not to be relied upon to dispose of plutonium feed stock. Introducing the plutonium to the yet-to-be-commissioned aqueous recovery line could reclaim future supplies, but purity of feedstock for the line is essential. The plutonium would have to be chemically cleaned, verified clean, dried, and stored beforehand. All that would be a costly addition of hazardous processing steps.

Doses to the maximally exposed worker from the two days of material handling were:

| | | | |
|---|---|---|---|
| Hand 1 | | | 0.77mSv |
| Hand 2 | | | 0.39 mSv |
| TLD, | Gamma | Shallow | 0.25 mSv |
| | Gamma | Deep | 0.25 mSv |
| | Neutron | | 0. |

This is well within the acceptable level of up to 0.79 mSv whole body dose predicted by an ALARA review. There was no observed change in the neutron or gamma flux when physically breaking and crushing the item. As predicted, the neutron and gamma dose rate to the worker is minimal once the material is in aqueous processing. Doses were incurred mostly while physically breaking up the material to prepare for aqueous processing, not during the processing itself. Because the dose was acceptable, the experience of Phase I led to the decision to use the physical break up method in Phase II and not the dissolution method.

**Approval for Disposal as Waste**

It is all very well to break up or dissolve and separate the materials, but the decision to throw the material away or reclaim it depends on factors beside technical feasibility; there are administrative issues. Is there a path forward for the waste? Can the material be discarded as is or will costly dilution or encapsulation be required for regulatory reasons? Is there a user for any reclaimed materials? Is there a capability for reclaiming or recycling the plutonium?

There would be a path forward for this material as waste; it could go to WIPP whether it was highly processed or simply hit with a hammer. In contrast there is currently no capability for reclaiming the material.

The Department of Energy (DOE) granted two key approvals for this project. They were expressed in two memoranda to LANL. One was for approval to dispose of specified attractiveness category D material[i] as waste. The second was for approval to dispose of these particular items. With these approvals in place, the project moved forward into Phase II.

## PHASE II: PHYSICAL BREAKUP AND REPACKAGING

The purpose of Phase II was to repackage ten items for disposal at WIPP as a proof of principle. Funding available was approximately $800k for combined Phase I experimentation and Phase II production. Later, additional funds provided for packaging nine more items.

**Location, Contamination, and Exposure Issues**

Determining where to conduct the packaging operation was a major concern. In Phase I we learned that the dose rates from crushed material did not rise to unacceptable levels, meaning that the work could be done in a glovebox without unduly irradiating the workers. Nevertheless, it was prudent to explore performing the operation in a hot cell in Wing 9 of the CMR building so that doses could be reduced to near zero. The hot cells provide superior radiation shielding and a large MAR limit (compared to other wings of the building) that would allow multiple items in the area at one time. However, the hot cells, even with a liner (an alpha box) to contain fine particles, would not provide sufficient control over the $^{238}$Pu oxide to solve the airborne contamination problem. Because of the spontaneous fragmentation[1] and extreme mobility of Pu-238 oxide, extraordinary control measures would be needed to minimize the possibility of a contamination release.

Based on prior experience with repackaging Pu-238 oxide waste[2] the realistic options for eliminating contamination issues in the hot cells were either to create an engineered containment structure within the alpha box or to use a glovebag inside the alpha box. Experienced hot cell manipulator operators counseled against using the glovebag inside a hot cell. Time and budget constraints prohibited the engineering solution, MAR limits in the CMR Building restricted the activity to a very inefficient level. All those factors made the CMR work location undesirable. But locating the operation at LANL's plutonium facility at TA-55 solved MAR limit issues and provided a glovebox where the work could be done, thus greatly reducing the contamination potential. A prepared glovebag (see Figure 1) was used inside a Pu-239 glovebox to minimize Pu-238 contamination to the glovebox interior and to reduce the likelihood of a contamination release from the glovebox.

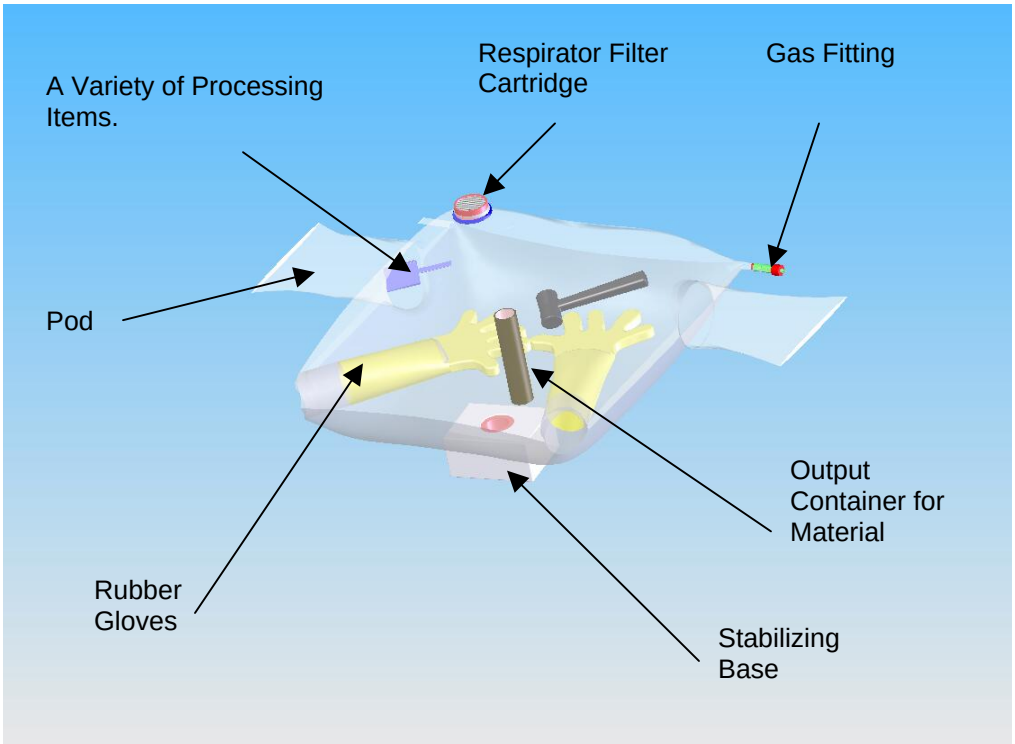


**Fig. 1. Prepared Glovebag for Use Inside a Glovebox**

## Glovebag Preparation

An $I^2R$ brand, model SS-30-20H, 3-mil polyethylene glovebag was modified by replacing the original gloves with more puncture resistant and better fitting natural rubber/neoprene/nitrile blend gloves. A gas fitting was taped into the supplied port to inflate the glovebag with inert gas, and a respirator filter cartridge was installed to vent the gas without releasing contamination. Through a hole cut in the bottom of the glovebag, a polyethylene cup was attached to hold the output container. The cup protected the bag from the forces of material crushing, extended the height of the bag, and fit into the stabilizing base. The stabilizing base remained outside the glovebag and provided stability and neutron shielding.

The bag had 2 pods, one closed and one open. The open pod was used for introduction of the output container (see Figure 2), the nuclear material, tools, and supplies (see Table I). The edges of the closed pod were reinforced with yellow vinyl tape to protect it when the output container was placed into it.

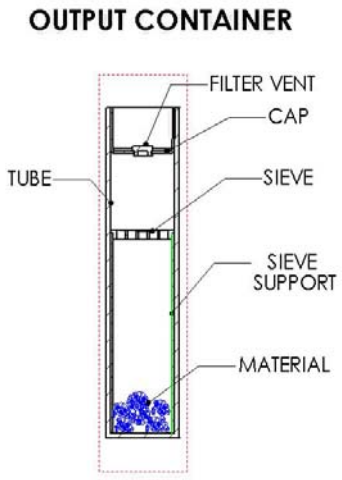


**Fig. 2. All-Metal Output Container for Waste**

**Table I. List of Materials Inside Bag**

| | |
|---|---|
| Shock absorbing pad pieces (rubber, 3 each) | Cleaning punch |
| | Damp decon cloths in sealed bags |
| Nuclear material | If required, can opener |
| Funnel | If required, channel locks |
| Output container with sieve and sieve support | Hammer |
| | Paint brush and scoop (use as strike plate if desired) |
| Cap with filtered vent | |
| Crusher with tape piece for cap | Tape cutter |

## Physical Breakup and Repackaging

The glovebag, tools, supplies, and material were introduced to the glovebox. The neutron emitter package and the required tools were introduced to the glovebag and the open pod was sealed with yellow vinyl tape and the glovebag was inflated to provide working room. The emitter packaging was opened and the material was removed and placed into a funnel positioned in the output container. The material was tamped through the funnel onto the top of a screen and then through the screen. Once the material was safely below the screen (and was incidentally shielded by the stabilizing base) a copper cap with a sintered metal filter was pounded into the output container creating a vented all-metal waste container. The output container cap was taped for extra security and the entire package wiped down with damp cheesecloth and placed into the reinforced pod for bagout.

The output container was removed from the glovebag (both still remaining inside the glovebox) by the twist, tape, and cut technique. The process waste was also twist-tape-and-cut removed from the glovebag for disposal. The tools were then isolated into the remaining bag and saved for introduction into the next item's glovebag. The output container was prepared for removal from the glovebox and subsequent calorimetry assay prior to being placed into a WIPP-approved 55-gallon drum Pipe Overpack Container (POC).[b] To date each POC contains only one item but experience may show that several items could be put into a single drum. The WIPP transportation limit of 2.0 mSv/hr at contact with the drum surface appears to be the limiting factor for packaging.

**Ergonomic Issues**

Using the glovebag put stress on worker's arms and hands beyond normal glovebox work. Workers wore surgical gloves and then worked in two additional sets of gloves, (glovebox and glovebag). The work included frequent bag cuts and taping inside the glovebox. This method also presented some additional stress when using a hammer, crusher, punch and strike plate. To ease the strain, the workers took frequent breaks and rotated the tasks between individuals so that the project was completed with no ergonomic injuries.

**Ionizing Radiation Issues**

Workers recorded gamma/beta and neutron readings of each original and final output container throughout the entire process (see Table II). Not unexpectedly the neutron dose rate increased after the crushing and sieving because of the increased Pu-238/beryllium interaction. Blocks of polyethylene shielding were used inside the glovebox to reduce the neutron exposure. The first ten items were repackaged in about four weeks. Assay, verification, and waste, operations were concluded about two months later. Worker doses were acceptable and there were no contamination incidents so the project was extended to work off nine more items. These additional items were processed using the same method as the first 10 items with similar results.

**Table II. Dose Rates of Material**

| | **ORIGINAL CONTAINER** (mSv/hr@ contact) | | **OUTPUT CONTAINER** (mSv/hr@ contact) | |
|---|---|---|---|---|
| | Beta-gamma | Neutron | Beta-gamma | Neutron |
| Item 1 | - | - | 0.22 | 1.01 |
| Item 2 | 0.17 | 0.16 | 0.19 | 0.10 |
| Item 3 | 0.14 | 0.21 | 0.26 | 0.84 |
| Item 4 | 0.27 | 0.46 | 0.10 | 0.50 |
| Item 5 | 0.12 | 0.17 | 0.23 | 0.17 |
| Item 6 | 0.27 | 0.48 | 0.10 | 0.51 |
| Item 7 | 0.09 | 0.20 | 0.11 | 0.38 |
| Item 8 | 0.15 | 0.17 | 0.08 | 0.17 |
| Item 9 | 0.23 | 0.39 | 0.35 | 0.54 |
| Item 10 | 0.16 | 0.18 | 0.22 | 0.16 |
| Item 11 | - | - | - | - |
| Item 12 | 0.10 | 0.93 | 0.17 | 0.23 |
| Item 13 | 0.005 | 0.009 | 0.005 | 0.01 |
| Item 14 | 0.09 | 0.61 | 0.18 | 1.20 |
| Item 15 | 0.23 | 0.12 | 0.06 | 0.07 |
| Item 16 | 0.16 | 0.15 | 0.48 | 1.20 |
| Item 17 | 0.17 | 0.19 | 0.50 | 0.60 |
| Item 18 | 0.015 | 0.26 | 0.16 | 0.65 |
| Item 19 | 0.07 | 0.27 | 0.15 | 0.66 |

To help control the total and individual doses, technicians wore supplemental alarming dosimetry in addition to a standard thermoluminescent dosimeter (TLD) on their body. The alarming dosimeter was set to notify at 0.25 mSv per shift (0.2 mSv neutron + .05 mSv gamma/beta). Stationary room monitors were placed in the room to provide real time readings of the radiation within the work area. Work would stop and be reevaluated if the alarms sounded. Standard dosimetry was worn on the wrist and chest. To avoid too much exposure to any person, workers rotated tasks among individuals. Total doses by individual for the final nine items are listed in Table III.

**Table III. Cumulative Doses to Individuals for Last 9 Items**

| | Shallow Dose | Deep Dose | Neutron Dose | Extremity | All dose from Supplemental |
|---|---|---|---|---|---|
| Worker 1 (TLD) | 0.23 mSv | 0.23 mSv | 0.45 mSv | 0.73 mSv (gamma) | |
| Worker 1 (supplemental) | | | | | 0.62 mSv |
| Worker 2 (TLD) | 0 | 0 | 0.22 mSv | 2.13 mSv (neutron) | |
| Worker 2 (supplemental) | | | | | 0.13 mSv |
| Observer 1 (TLD) | 0.22 mSv | 0.22 mSv | 0.44 mSv | 0.78 mSv (gamma) | |
| Observer 1 (supplemental) | | | | | 0.24 mSv |
| Observer 2 (TLD) | 0 | 0 | 0 | 0 | |
| Observer 2 (supplemental) | | | | | 0.03 mSv |
| Observer 3 (TLD) | 0.13 mSv | 0.13 mSv | 0.20 mSv | | |
| Observer 3 (supplemental) | | | | | 0.07 mSv |
| Observer 4 (TLD) | 0 | 0 | 0.11 mSv | 0.21 mSv (gamma) 0.21 mSv (neutron) | |
| Observer 4 (supplemental) | | | | | 0.02 mSv |
| **Total of all Workers** (TLD) | 0.58 mSv | 0.58 mSv | 1.42 mSv | 1.87 mSv (gamma) 2.49 mSv (neutron) | |
| **Total of all Workers** (supplemental) | | | | | 1.11 person-mSv |

The best measurement of the dose is from supplemental dosimetry and is 1.11 person-mSv for nine items. A conservative estimate of the dose to the maximally exposed individual would be no less than 0.62 mSv per nine items or 0.07 mSv per person for each item.

A less accurate measurement of dose is had from the TLDs. By that measure the total dose for nine items was 2.58 person-mSv for nine items. The dose to the maximally exposed individual would be 0.91 mSv for nine items or 0.1 mSv per item per person.

Prior to performing the work, the total expected dose was estimated to be 5.8 person- mSv based on the repackaging process and nearby processes in the work area. Actual doses were much less than estimated because the co-located activities were not being conducted at the time.

**Contamination Issues**

Swipe surveys taken inside the glovebox showed a gross alpha contamination of millions of counts per second per 100 $cm^2$. Such swipes were too hot to send to the analysis laboratory so derivative swipes were taken (swipes taken from the original swipes) and counted. The swipes indicated no detectable Pu-238 activity. The lack of Pu-238 activity is testimony to the effectiveness of glovebags to contain contamination.

**Waste Generation**

Several episodes of handling are required between glovebox bagout and final placement into a POC. Careful examination of the process to date, concentrating on ALARA principles, has led to placing only a single item in each POC. Therefore the 19 items at 206 liters each represent 3.91 cubic meters of TRU waste. Incidental waste such as original packaging, tools, handling supplies and decontamination waste accounted for an additional 1.03 cubic meters of TRU waste. Total waste generation including decontamination trash was as follows:

1) 19 items, one item per drum----- 3.91 $m^3$ TRU

2) Metal waste----------0.41 $m^3$ TRU

3) Plastic waste----------0.41 $m^3$ TRU

4) Combustible Waste----------0.21$m^3$ TRU

TOTAL WASTE---------- ~5$m^3$ TRU.

**COST AND SCHEDULE**

The total time for packaging, bagging out, assaying, and transporting the 19 items and ancillary waste was approximately 4 months.

Phase I – open and examine one item; determine likely doses and plan the path forward; analyze samples and dispose of waste $385k

Phase II – process the first ten items; manage process waste $406k

Phase III (added based on the success of Phase II) – process the next nine items and clean out the glovebox for the next user $400k

PROJECT TOTAL COST **$1,191k**

## RESULTS

As seen in Table III, the neutron dose rate from the material increased by up to a factor of five during repackaging. This was not unexpected. The maximum dose rate of 1.68 mSv/hr at contact for an item did not exceed the safe handling level of 0.50 mSv/hr at the work distance identified in the work authorizing documents. The first 10 items were successfully repackaged with the physical break up method so an additional 9 items were added to the work scope. There were no radioactive releases to the room. The total exposure to personnel was 1.11 person-mSv. A safe estimate for the maximally exposed worker is 0.07 mSv per item per person. At that rate, a team could package over 700 items in one year without exceeding the LANL dose limit of 50 mSv per person in a year.

The glovebox was returned to Pu-239 service based on the contamination survey data. Planning and experimentation took nearly two years but the repackaging time was about four months for a total project cost of about $1.2M. The cost and schedule included glovebox clean up and waste disposal.

The neutron emitters did not increase in dose rate beyond 1.68 mSv /hr at contact when physically broken into small pieces, allaying fears that spikes in dose rate would prevent the handling of material in this fashion. The total volume of WIPP-certifiable waste generated was 4.9 cubic meters. The crushing/sieving method proved to be a safe and economical solution for these legacy waste items. The data collected for the first 10 items provided support for extending the project to an additional 9 items. The cumulative personnel radiation dose was well within limits and actually proved to be lower than anticipated for the second 9 items. The use of glovebags was a totally effective defense in depth to prevent a contamination release to the room.

## CONCLUSION

The items can be safely packaged for disposal at WIPP by using a physical breakup method. A chemical dissolution and separation process works but does not solve any significant problems of the process and it creates additional problems that the physical break up method does not. Glovebags can be considered when facing the challenge of working with highly dispersible materials in an environment where contamination is unacceptable.

Future work in this area includes the working off any remaining materials of this kind. Packaging efficiency might be gained if more than one item could be placed into a WIPP-compliant output container or if several items could be placed into a single 55-gallon drum. To help alleviate the additional stress on the technicians arms and hands due to working in a glovebag inside a glovebox, a glovebox that is specifically dedicated for Pu-238, or a specially designed containment box inside the Pu-239 glovebox should be considered.

## REFERENCES

Clinard, F.W. Jr. and Rohr, E. L., "Spontaneous Fragmentation of an Alpha-Active Ceramic – A Mechanism for Dispersion of Solid Waste?" *Scientific Basis for Nuclear Waste Management*, Vol. 3, pp. 405 - 412, John G. Moore, ed. Plenum Publishing Corporation, 1981.

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## FOOTNOTES

1. DOE M474.1 defines Category E as 1gm SNM/Kg net of contaminated/impure oxides. Material in this project was more concentrated than that.
2. A Pipe Overpack Container (POC) is an all-metal container with a bolted lid that fits inside a padded 55-gallon DOT Type-A drum. The design intent is to increase the amount of non-hydrogenous waste that can be shipped to WIPP in a 55-gallon drum. The limiting factor for these items in a POC appears to be the external dose rate of < 2.0 mSv/hr at contact, followed by a volume limit that depends on how much protective material is needed around an item to safeguard operators during assay, packaging, and other handling.