

# DISPOSAL OF <sup>233</sup>U IN THE WASTE ISOLATION PILOT PLANT

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## DISPOSAL OF $^{233}\text{U}$ IN THE WASTE ISOLATION PILOT PLANT

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### ABSTRACT

The United States is considering the disposal of excess uranium-233 ( $^{233}\text{U}$ ), which is a weapons-usable, alpha-emitter with many properties similar to those of plutonium (Pu). Seven processing options were identified to convert the  $^{233}\text{U}$  into a form acceptable for disposal into the Waste Isolation Pilot Plant (WIPP). Processing is required to meet two WIPP waste acceptance criteria (WAC): (a) criticality control and (b) safeguards. There are multiple approaches to meet these criteria. Uranium-233 can be converted to non-weapons-usable  $^{233}\text{U}$  by mass dilution with transuranic waste (TRUW) or by isotopically diluting the  $^{233}\text{U}$  with  $^{238}\text{U}$  (<12 wt %  $^{233}\text{U}$  in  $^{238}\text{U}$ ). Uranium-233 can be made critically safe by the limiting fissile mass per drum, adding gadolinium, or isotopically diluting  $^{233}\text{U}$  with  $^{238}\text{U}$  (<0.66 wt % in  $^{238}\text{U}$ ). The isotopic dilution options would require changes in the current WAC. Isotopic dilution techniques were not considered when the WIPP WAC was originally developed because the expected concentrations of  $^{233}\text{U}$  in waste streams at that time were very low. There are also legal uncertainties. WIPP accepts only defense-related TRUW. Some—but not necessarily all—of the  $^{233}\text{U}$  contains sufficient plutonium such as to be considered TRUW if it were declared waste.

### INTRODUCTION

The United States is considering the disposition of excess  $^{233}\text{U}$ . Plutonium-239,  $^{235}\text{U}$ , and  $^{233}\text{U}$  are the three weapons-usable fissile isotopes that can be produced in large quantities. All have been investigated for use in weapons, naval reactors, and commercial nuclear power reactors. For a variety of technical, institutional, and economic reasons,  $^{233}\text{U}$  is not currently used for any of these applications. Furthermore, much of the inventory (1) consists of  $^{233}\text{U}$  mixed with other uranium isotopes, which thus limits potential future uses (Table 1). The United States is considering disposition of some  $^{233}\text{U}$  based on three factors: (a) storage costs, (b) limited near-term needs, and (c) arms control. With the end of the cold war, the United States and Russia are simultaneously reducing their inventories of weapons-usable materials. No  $^{233}\text{U}$  disposition decisions have been made at this time.

Uranium-233 can be processed before its disposal for the recovery of thorium-229 ( $^{229}\text{Th}$ )—a decay product of  $^{233}\text{U}$ . Thorium-229 is the precursor used to produce bismuth-213 ( $^{213}\text{Bi}$ ), which is being investigated in clinical trials to determine the effectiveness of  $^{213}\text{Bi}$  for treating certain cancers. There are several potential methods to produce  $^{213}\text{Bi}$ ; however, the  $^{229}\text{Th}$  derived from  $^{233}\text{U}$  is the only currently available source of this medical isotope. If  $^{213}\text{Bi}$  becomes a standard approach for cancer treatment, large quantities of  $^{213}\text{Bi}$  would be required. Since only limited amounts of  $^{213}\text{Bi}$  are available from  $^{233}\text{U}$ , alternative methods to produce  $^{213}\text{Bi}$  would be implemented. It takes 10 to 20 years for a significant amount of  $^{229}\text{Th}$  to build up in  $^{233}\text{U}$ ; thus, there may be incentives to first recover the medical isotopes from  $^{233}\text{U}$  and then consider the  $^{233}\text{U}$  as a waste. Such operations may generate significant additional quantities of high-alpha wastes.

**Table I. Major batches of separated <sup>233</sup>U in inventory<sup>a</sup>**

Batch No.	Location/designation	Material and packaging	Total U (kg)	Uranium isotopics		
				<sup>233</sup> U (kg)	<sup>235</sup> U (kg)	<sup>232</sup> U (ppm) <sup>a</sup>
<i>High isotopic quality with limited chemical impurities</i>						
1	Oak Ridge National Laboratory (ORNL)	U <sub>3</sub> O <sub>8</sub> monolith in 27 welded stainless steel cans	65.2	60.3	0.0	15
2	ORNL (2 similar batches)	UO <sub>x</sub> powder in 247 cans	108.8	103.1	0.0	4–9
3	ORNL	U <sub>3</sub> O <sub>8</sub> powder	46.0	45	0.0	6
4	Multiple/remaining small lots	Many forms and packages	49.0	47.9	- 0.0	
Subtotal			269.0	256.3		
<i>High isotopic quality with chemical diluents (ThO<sub>2</sub> or ZrO<sub>2</sub>)</i>						
5a <sup>b,c</sup>	Idaho National Environmental and Engineering Laboratory (INEEL)/Light-Water Breeder Reactor (LWBR)	Unirradiated rods and pellets with 483 kg ThO <sub>2</sub>	29.5	28.5	0.0	9
5b	INEEL/LWBR (ZrO <sub>2</sub> )	Unirradiated rods and pellets made of <sup>223</sup> UO <sub>2</sub> and ZrO <sub>2</sub>	5.6	5.5	0.0	38
6 <sup>b</sup>	INEEL/LWBR	Unirradiated LWBR fuel with 14 t natural thorium	323.5	317.4	0.0	9
Subtotal			358.6	351.4		
<i>Intermediate isotopic quality</i>						
7	ORNL/Savannah River Site	UO <sub>3</sub> powder in 140 cans	67.4	61.6	0.0	156
8 <sup>e</sup>	ORNL/Molten Salt Reactor Experiment (MSRE) <sup>d</sup>	UO <sub>x</sub> powder after conversion	40.6	33.9		160–200
Subtotal			108.0	95.5		
<i>Low isotopic quality</i>						
9	ORNL/Consolidated Edison Uranium Solidification Program (CEUSP)	U <sub>3</sub> O <sub>8</sub> monolith in 403 cans	1,042.6	101.1	796.3	120
10	Clean/Y-12	UO <sub>x</sub> powder in 5 cans	42.6	0.9	38.7	6
Subtotal			1,085.2	102.0		
Total			1,820.8	805.3		

<sup>a</sup>Based on the uranium content.

<sup>b</sup>The uranium is in the form of UO<sub>2</sub>-ThO<sub>2</sub> fuel pellets with 1 to 10 wt % <sup>233</sup>U. The average assay is - 2 wt % <sup>233</sup>U.

<sup>c</sup>One drum of 188 g <sup>233</sup>U metal in 9.3 kg thorium metal.

<sup>d</sup>Material in inventory and being recovered from the MSRE. The material will be converted to oxide form for storage.

Uranium-233 is an alpha emitter. It must be diluted by  $10^5$  before the alpha activity approaches 100 nCi/g of  $^{233}\text{U}$ —the generally accepted concentration boundary between shallow-land and geological disposal of alpha wastes. As a consequence, protection of human health requires disposal of  $^{233}\text{U}$  wastes in a deep geological repository—such as WIPP or the proposed Yucca Mountain (YM) repository. This evaluation was conducted to identify and evaluate potential methods to process the  $^{233}\text{U}$  for acceptance by WIPP.

## PROPERTIES OF $^{233}\text{U}$

Uranium-233 has many properties similar to those of Pu. The quantity of  $^{233}\text{U}$  necessary to build a nuclear weapon, as defined by the International Atomic Energy Agency, is the same as that for plutonium. The critical masses are similar. Both are alpha emitters; the alpha activity of  $^{233}\text{U}$  is somewhat less than that of  $^{239}\text{Pu}$ . The half-life of  $^{239}\text{Pu}$  is 24,000 years, whereas the half-life of  $^{233}\text{U}$  is 160,000 years. Uranium-233 is always processed in gloveboxes or hot cells.

Uranium-233 has some properties (2,3) that are different from those of plutonium. Uranium-233 can be isotopically diluted with  $^{238}\text{U}$  to (a) convert it to non-weapons-usable  $^{233}\text{U}$  [ $<12$  wt %  $^{233}\text{U}$  in  $^{238}\text{U}$ ] and (b) prevent the possibility of nuclear criticality [ $<0.66$  wt %  $^{233}\text{U}$  in  $^{238}\text{U}$ ]. Plutonium can not be isotopically diluted to eliminate safeguards and criticality concerns.

Uranium-233 contains varying concentrations of  $^{232}\text{U}$  and its gamma-emitting decay product ( $^{208}\text{Tl}$ ). Because of this  $^{232}\text{U}$  decay product, much of the  $^{233}\text{U}$  requires radiation shielding for safe handling and storage. Figure 1 shows the radiation levels from a canister containing 1 kg of  $^{233}\text{U}$  and 100 ppm  $^{232}\text{U}$  as a function of time. The inventory (3) contains different lots of  $^{233}\text{U}$  with different concentrations of  $^{232}\text{U}$ . About half the inventory has  $^{232}\text{U}$  concentrations near 100 ppm  $^{232}\text{U}$ . The remainder of the inventory has much lower  $^{232}\text{U}$  concentrations.

From a technical perspective, WIPP is a logical disposal facility for any  $^{233}\text{U}$  waste. WIPP is designed as an alpha repository. Uranium-233 is an alpha-emitting material, which is similar to that of plutonium. The hazards and properties of the two materials are similar and thus require disposal facilities with the same functional requirements and capabilities

## WIPP CONSTRAINTS

The WIPP facility (4) has defined WAC. *Wastes* are defined as either contact-handled (CH) or remote-handled (RH). A package is CH waste if the external radiation level at the surface of the drum is  $<200$  mrem/h. If the radiation levels are higher, it is RH waste. However, the radiation level may not exceed 1,000 rems per hour. Because of the radiation dose from the  $^{232}\text{U}$  impurity in  $^{233}\text{U}$ , a waste drum may be either CH or RH, depending upon the  $^{232}\text{U}$  content. Other important WAC are shown in Fig. 2. Risks from equipment failure, accidents, and nuclear criticality are controlled by placing limits on (a) drum weight, (b) waste form and radionuclide inventory, and (c) fissile mass.

WIPP currently accepts only post-1970, defense TRUW. While  $^{233}\text{U}$  is not a transuranic element, some existing TRUW contain  $^{233}\text{U}$ . As a consequence of these existing wastes that will be disposed of in WIPP, (a) the performance assessments for WIPP considered  $^{233}\text{U}$  and found it was acceptable to disposal of  $^{233}\text{U}$  in WIPP, (b) the permits and other licensing documents allow  $^{233}\text{U}$  into WIPP, and (c) WIPP WAC explicitly define  $^{233}\text{U}$  disposal requirements. There are however several constraints.

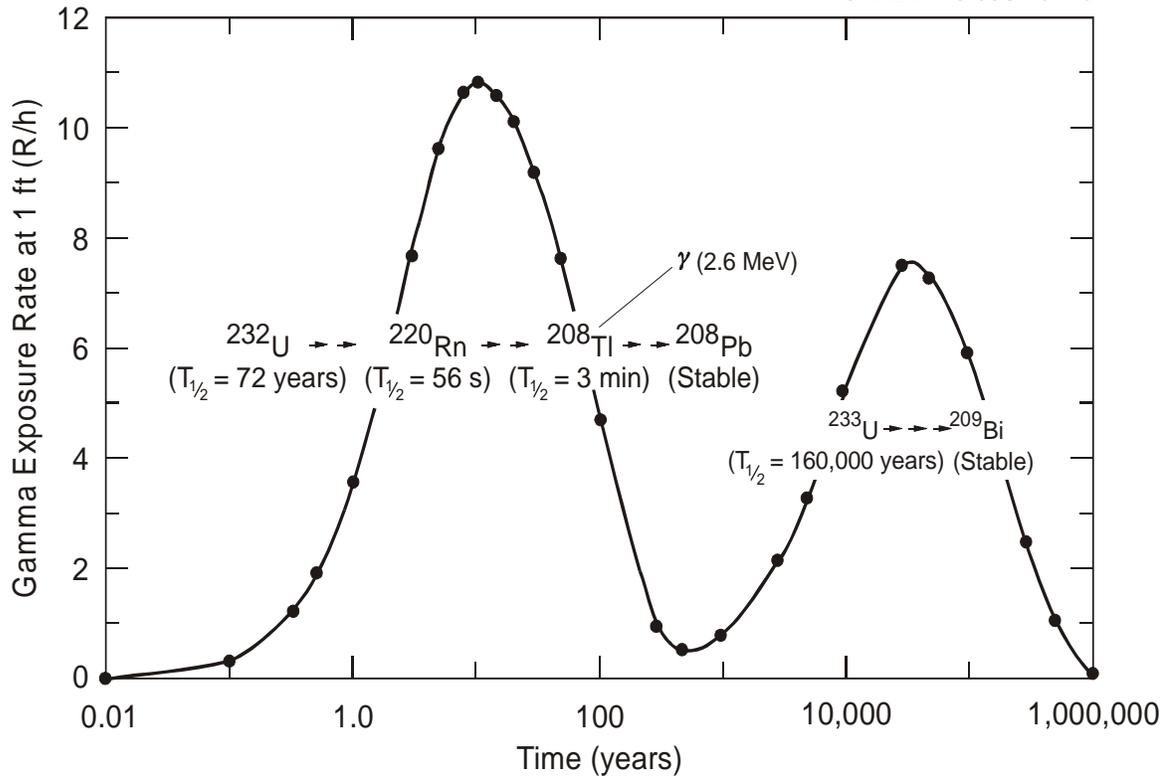


Fig. 1. Gamma exposure for 1 kg  $^{233}\text{U}$  with 100 ppm  $^{232}\text{U}$ .

## Legal

The legal authorization basis in the *WIPP Land Withdrawal Act* limits the use of WIPP for the disposal of post-1970 defense TRUW. Uranium-233 is not defined as a TRU isotope although historically it has been managed as such at U.S. Department of Energy sites. Much—but not necessarily all—of the  $^{233}\text{U}$  in storage contains sufficient Pu ( $\sim 1$  ppm) such that it would be TRUW if it were declared a waste. Much—but not necessarily all—of the  $^{233}\text{U}$  in storage has been associated with defense programs. Some fraction of the  $^{233}\text{U}$  inventory may not meet the legal requirements for acceptance into WIPP.

The same facilities were often used to process Pu and  $^{233}\text{U}$  because  $^{233}\text{U}$  is (a) an alpha emitter and requires similar handling techniques as those used for Pu (gloveboxes or hot cells), (b) the criticality control limits are usually identical to those of Pu, and (c) the safeguards requirements are identical. These processes resulted in some Pu contamination of the  $^{233}\text{U}$ . For most applications, there were (and still are) no penalties if the  $^{233}\text{U}$  contained (or contains) small quantities of plutonium. Therefore, there was no reason to conduct expensive operations to remove low levels of plutonium (up to a few hundred parts per million of plutonium) from the  $^{233}\text{U}$ . As a consequence, much of the  $^{233}\text{U}$ , if declared a waste, would be classified as TRUW.

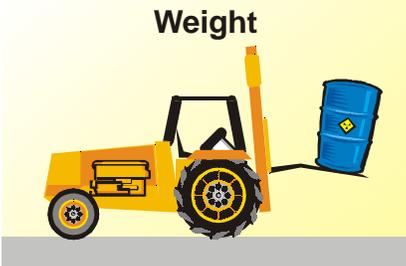
Criteria	TRU Waste Category	
	Contact Handled (55-gal Drum)	Remote Handled (RH Container Accepts 3 Drums)
<p><b>Weight</b></p> 	≤450 kg	≤3600 kg
<p><b>Accident Consequence Control (Radioactivity)</b></p> 	<p>Untreated Waste ≤1.3 kg <sup>233</sup>U</p> <p>Treated Waste ≤29 kg <sup>233</sup>U</p>	<p>≤16 kg <sup>233</sup>U</p> <p>≤16 kg <sup>233</sup>U</p>
<p><b>Criticality</b></p> 	<p>Fissile Mass Limit ≤200 g <sup>233</sup>U/ Drum</p> <p>Isotopic Dilution Limit No Limit</p>	<p>≤325 g <sup>233</sup>U/ RH Container</p> <p>No Limit</p>

Fig. 2. Technical WIPP WAC constraints.

There is a second legal issue. While the concentrations of Pu in the  $^{233}\text{U}$  are sufficient to make most of the  $^{233}\text{U}$  in inventory a transuranic material, any processing will produce secondary processing wastes [high-efficiency particulate air (HEPA) filters, equipment, etc], which will contain lower Pu concentrations below the concentration threshold defined for TRUW—100 nCi/g of transuranic elements. At the same time, the  $^{233}\text{U}$  concentrations will be far above 100 nCi/g of alpha activity and thus not suitable for shallow-land disposal. In addition, there are - 2500 drums (5) of  $^{233}\text{U}$  orphan wastes with  $^{233}\text{U}$  concentrations above 100 nCi/g of alpha activity.

The disposal of high-alpha wastes that are not defense TRUWs is an unresolved policy issue. Options for the disposal of these materials are: (a) change federal law to allow WIPP to accept other alpha wastes with characteristics similar to those of TRUWs, (b) dispose of these wastes in the proposed YM repository, or (c) construct a new repository for the very-small quantities of orphan alpha wastes.

While the YM repository is an option, it is designed for spent nuclear fuel and high-level waste that generate significant quantities of decay heat. Because of the resultant elevated temperatures and other considerations, the proposed YM repository prohibits organics in any waste stream and impose several other restrictions on the waste form. To meet the YM WAC would require new processing facilities to convert existing and future  $^{233}\text{U}$  wastes into an acceptable waste form. Repositories designed for different types of waste have different WAC, which in turn impose different requirements on the waste forms. For this analysis, it is assumed that WIPP can legally accept all wastes containing  $^{233}\text{U}$ .

## **Nuclear Criticality**

The WIPP facility controls nuclear criticality by limiting the equivalent fissile content of each container. A single, CH, 208-liter (55-gal) waste drum is limited to <200 g of  $^{233}\text{U}$  if  $^{233}\text{U}$  is the only fissile material. The mass limits are different for different fissile isotopes. A sum-of-the-fraction mixture rule determines the quantity of fissile materials which can be put into a drum if there is more than one fissile material in the waste stream. If the waste is packaged in a large (equivalent to three 208 liter drums) RH waste package (WP), the limit is expected to be 325 g  $^{233}\text{U}$  equivalent per container.

Alternatively, the  $^{233}\text{U}$  can be isotopically diluted with depleted uranium (DU). This second approach to criticality control is not in the current WIPP WAC because, when these limits were being developed, there was no consideration of disposing of  $^{233}\text{U}$  in high concentrations. Fissile materials were considered valuable and thus were not expected in significant concentrations in any waste stream.

Isotopic dilution is a more conservative approach to nuclear criticality control than mass limits on individual WPs. To ensure criticality control under all conditions using isotopic dilution (3),  $^{233}\text{U}$  must be diluted to - 0.66 wt %  $^{233}\text{U}$  in  $^{238}\text{U}$ . This is equivalent to - 1 wt %  $^{235}\text{U}$  in  $^{238}\text{U}$ . If  $^{233}\text{U}$  is isotopically down-blended, the DU will contain some  $^{235}\text{U}$ . Some of the  $^{238}\text{U}$  in the DU is necessary to maintain criticality control of the  $^{235}\text{U}$ . Consequently, the final blend would typically contain - 0.5 wt %  $^{233}\text{U}$ , a small quantity of  $^{235}\text{U}$  from the DU, and the  $^{238}\text{U}$ . If isotopic dilution is used, the  $^{233}\text{U}$  content of a CH drum can be increased from about 200 g (fissile drum limit) to about 2000 g. Package weight limits (450 kg for a CH drum) control the total quantity of  $^{233}\text{U}$  and DU per drum. This criticality approach can reduce the number of WPs by a factor of 10.

## Safeguards

WIPP does not accept weapons-usable materials. The facility has only industrial security. The security systems are designed to minimize theft of property (computers, tools, etc.). The transportation system has the same restrictions. Uranium-233 is weapons usable. Before it is sent to WIPP, the  $^{233}\text{U}$  must be converted to a non-weapons-usable material. There are two ways to accomplish this task (5).

- *Isotopic dilution.* The  $^{233}\text{U}$  can be isotopically diluted to <12 wt %  $^{233}\text{U}$  in  $^{238}\text{U}$ . The 12 wt % concentration in terms of the potential to build nuclear weapons is equivalent to the 20 wt %  $^{235}\text{U}$  in  $^{238}\text{U}$  that divides weapons-usable and non-weapons-usable  $^{235}\text{U}$ .
- *Mass dilution.* If the  $^{233}\text{U}$  is well mixed with sufficient waste, it can be declared non-weapons usable. There is no definition for *well mixed*. Presumably, grinding the  $^{233}\text{U}$  with the waste to a fine consistency would mix the material well enough to meet this requirement.

Technical studies (2) have defined non-weapons-usable  $^{233}\text{U}$ ; however, these limits have not been implemented into the safeguards regulatory structure. Until recently, there was no consideration of down-blending  $^{233}\text{U}$  and thus no definition of *non-weapons usable* was formally adopted.

## PROCESSING AND DISPOSAL OPTIONS

Seven options have been identified for processing  $^{233}\text{U}$  into a form that would be expected to be acceptable for disposal in WIPP. The processing and disposal options are shown in Fig 3.

### **Option 1: Co-Process Weapons-Usable $^{233}\text{U}$ With CH TRUW at the INEEL Advanced Mixed Waste Treatment Facility (AMWTF) and Ship to WIPP**

The AMWTF is currently under construction at INEEL to treat and repackage CH TRUW for shipment and disposal at WIPP. Wastes are treated and packaged to meet WIPP WAC (4). Treatment operations within the facility include (a) size-reduction of large components; (b) waste compaction; (c) solidification of liquids, sludges, and powders by mixing with cement and forming a cement waste product; and (d) other treatments. The initial goal is to process, package, and ship to WIPP the CH TRUW at INEEL. However, there is agreement with the State of Idaho that other wastes can be brought into this facility for treatment and packaging—provided that the processed wastes are shipped to WIPP within a specified period of time. This  $^{233}\text{U}$  treatment option consists of the following steps.

- *Shipment to INEEL.* The  $^{233}\text{U}$  is shipped to INEEL as weapons-usable material with appropriate security.
- *Grinding.* At INEEL, the  $^{233}\text{U}$  is ground into a fine powder and stored in high-security vaults.
- *Mixing.* The ground  $^{233}\text{U}$  is shipped from the high-security vaults to the AMWTF in small quantities. This avoids the need for weapons-type security requirements at the AMWTF. The  $^{233}\text{U}$  powder is then mixed with CH TRUW, and this mixture is packaged. One processing option within the facility is to simultaneously mix (a) the  $^{233}\text{U}$  powder; (b) TRUW sludges, liquids, and powders; (c) cement; and (d) additives to produce a qualified cement waste form for shipment and disposal in WIPP.

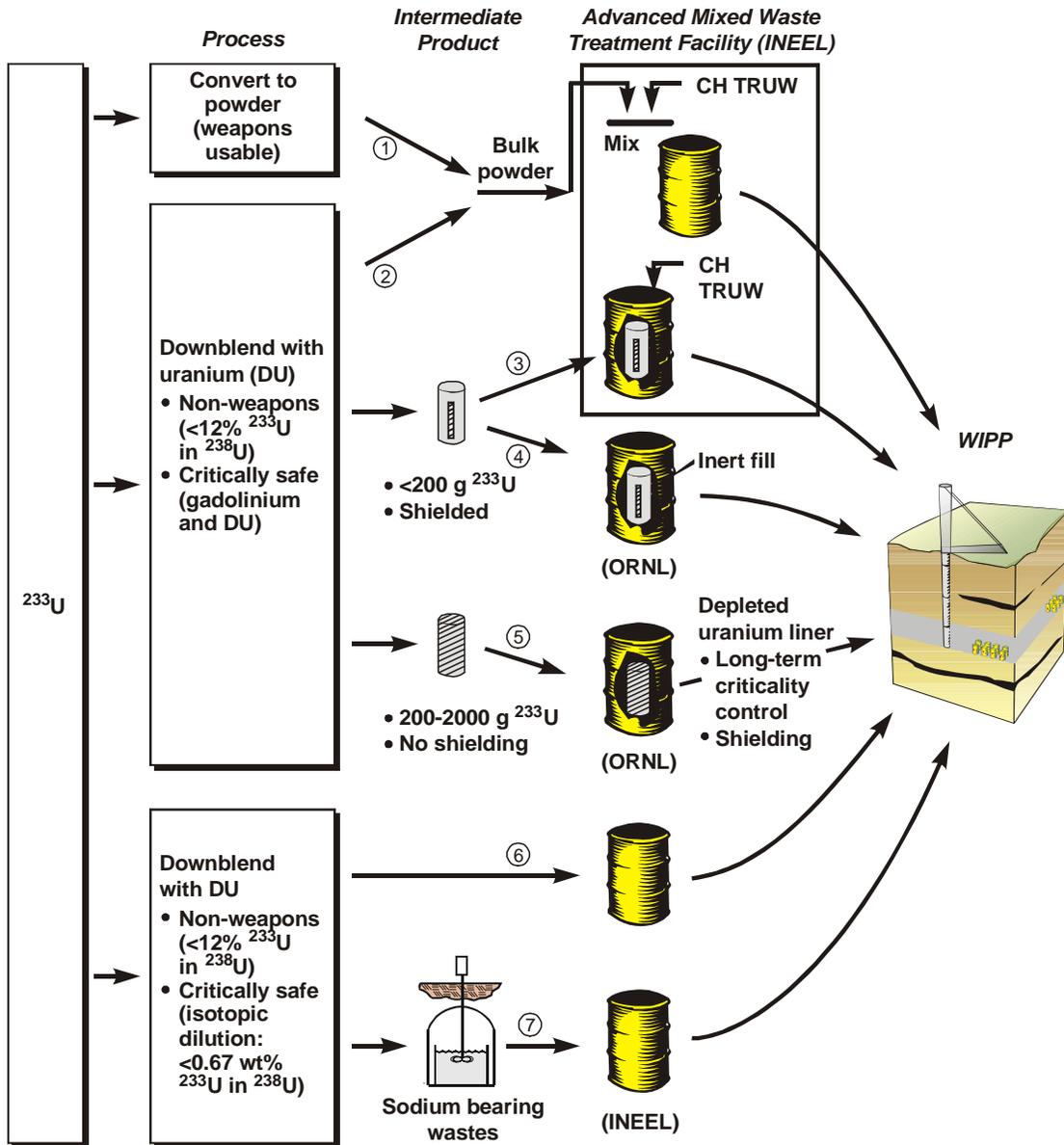


Fig. 3. WIPP  $^{233}\text{U}$  processing and disposition options.

- *Shipment from INEEL and disposal.* The CH TRUW with the  $^{233}\text{U}$  is shipped to and disposed of at WIPP.

The viability of this option is based on one characteristic of the AMWTF: This facility will process very large quantities of TRUW. In the initial Phase I campaign, it is expected that 36,530 m<sup>3</sup> of TRUW (equivalent to ~ 180,000 drums) will be processed (6). The average Pu content per drum is estimated at <4 g/drum. Because of the very large volumes of TRUW to be processed, only a few grams of  $^{233}\text{U}$  would be added to each drum of TRUW to dispose of the entire inventory. The quantity of  $^{233}\text{U}$  is far below the WIPP WAC criticality limit of 200 g of  $^{233}\text{U}$  equivalent per drum. In theory, several tens of tons of  $^{233}\text{U}$  could be mixed with this waste and still meet the WIPP WAC. The criticality control requirements limit the total quantities of fissile materials per drum; thus, if other fissile materials ( $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , etc.) are in the drum, the allowable quantity of  $^{233}\text{U}$  is reduced. The extreme dilution of the  $^{233}\text{U}$  with TRUW makes the  $^{233}\text{U}$  practically unrecoverable and thus non-weapons usable. The extreme dilution ensures that the radiation doses from the  $^{232}\text{U}$  in the  $^{233}\text{U}$  are acceptably low.

**Option 2: Isotopically Dilute  $^{233}\text{U}$  with  $^{238}\text{U}$  to Non-Weapons-Usable  $^{233}\text{U}$ , Mix with CH TRUW at the AMWTF, and Ship to WIPP**

This option is similar to the first option—except that initially the  $^{233}\text{U}$  is mixed on a molecular scale with  $^{238}\text{U}$  and gadolinium. After mixing, the product is sent to INEEL to be blended with TRUW at the AMWTF. The  $^{233}\text{U}$  is down-blended with DU to convert the  $^{233}\text{U}$  to non-weapons-usable  $^{233}\text{U}$  (<12 wt %  $^{233}\text{U}$  in  $^{238}\text{U}$ ). Isotopic dilution provides a clearly acceptable way to convert weapons-usable uranium isotopes to non-weapons-usable materials. The addition of gadolinium would be expected to eliminate operational nuclear criticality concerns.

The most likely mixing process would be the full dissolution of the  $^{233}\text{U}$ ,  $^{238}\text{U}$ , and gadolinium in nitric acid. The resultant homogeneous solution would be calcined to an oxide solid. This processing eliminates (a) security requirements for all subsequent operations, (b) assures that the product is non-weapons-usable, and (c) minimizes operational criticality issues. The same process is used to make nuclear fuels with gadolinium as a burnable neutron absorber. Consequently, there is a very-large experience base using gadolinium for criticality control in this system.

It is currently planned to recover  $^{229}\text{Th}$  from some batches of  $^{233}\text{U}$  for medical purposes. The standard method for recovery of this isotope is to dissolve the  $^{233}\text{U}$  in nitric acid and then remove the thorium by ion-exchange and solidification of the resultant uranium solution. If it were decided that the  $^{233}\text{U}$  was a waste after thorium recovery, the DU would be added to the nitric solution immediately after the ion-exchange process and before uranium solidification. This processing option and all subsequent options herein allow the recovery of medical isotopes from the  $^{233}\text{U}$ .

**Option 3: Isotopically Dilute  $^{233}\text{U}$  with  $^{238}\text{U}$  to Non-Weapons-Usable  $^{233}\text{U}$ , Place in a Shielded Minipackage, Co-Package with Other CH TRUW, and Ship to WIPP**

This option consists of the following steps.

- *Isotopic dilution.* The  $^{233}\text{U}$  is mixed on a molecular scale with  $^{238}\text{U}$  and gadolinium—as described above. The product is a non-weapons-usable (<12 wt %  $^{233}\text{U}$  in  $^{238}\text{U}$ ), critically safe solid. The oxide mixture is packaged in small, self-shielded packages with <200 g of  $^{233}\text{U}$  per minipackage.

- *Shipment.* The shielded  $^{233}\text{U}$ - $^{238}\text{U}$ -Gd oxide minipackages are shipped directly to the AMWTF as non-weapons-usable materials. The conversion of the  $^{233}\text{U}$  to non-weapons-usable  $^{233}\text{U}$  in self-shielded packages reduces shipping costs by minimizing security requirements and packaging requirements.
- *Addition.* One minipackage with <200 g of  $^{233}\text{U}$  equivalent in the form of a  $^{233}\text{U}$ - $^{238}\text{U}$ -Gd oxide is placed in a TRUW drum, and the empty space in the drum is filled with TRUW. The WIPP criticality control requirements limit the total quantities of fissile materials per drum; thus, if other fissile materials ( $^{235}\text{U}$ ,  $^{239}\text{Pu}$ , etc.) are in the drum, the allowable quantity of  $^{233}\text{U}$  is reduced. The drum is CH.
- *Shipment to and disposal at WIPP.* The CH TRUW with the  $^{233}\text{U}$  minicontainer is shipped to and disposed of in WIPP.

The  $^{233}\text{U}$  is non-weapons usable by isotopic dilution. The WIPP WAC criticality limit is met by limiting the  $^{233}\text{U}$  to <200 g equivalent per drum. Efficient use of waste drums is achieved by adding  $^{233}\text{U}$  minipackages to TRUW drums. The WIPP WAC limit the mass, volume, and fissile material content of a drum. The waste content of a typical drum is volume limited because of the low density of most TRUW (clothing, gloves, etc.). The typical fissile content of TRUW is also very low—usually only a few grams per drum and far below the fissile material limit of 200 g  $^{239}\text{Pu}$  equivalent per drum. The drum weight is far below the 450-kg drum mass limit. Adding a  $^{233}\text{U}$  minipackage consumes only a small volume fraction of the drum, but allows the efficient use of the fissile disposal limits of the drum. The rest of the void space is used for other TRUW. The low density of most TRUW allows the addition of a shielded minipackage without exceeded drum mass limits.

The use of shielded minipackages reduces occupational radiation exposures. Options 1 and 2 minimize radiation levels by mass dilution; however, this increases the radiation levels of many drums. Radiation exposure is spread over a larger number of people, but total exposure may or may not be reduced. It is highly dependent upon where the  $^{233}\text{U}$  is placed in each drum (center or near the outer edge of the package). The minipackage options use shielding to minimize total radiation exposure. WIPP currently accepts several packages with internal structures to minimize occupational hazards. This includes the pipe-and-go package for certain plutonium residues and other planned packages for other wastes with unique characteristics (7).

**Option 4: Isotopically Dilute  $^{233}\text{U}$  with  $^{238}\text{U}$  to Non-Weapons-Usable- $^{233}\text{U}$ , Place in Shielded Minipackage, Place with Inert Fill in Drum, and Send to WIPP**

This option is identical to Option 3—except the minipackages are placed in WIPP drums with added fill materials. No extra TRUW is added. This simplifies operations at the cost of producing additional waste drums that require disposal.

**Option 5: Isotopically Dilute  $^{233}\text{U}$  with  $^{238}\text{U}$  to Non-Weapons-Usable  $^{233}\text{U}$ , Place in Shielded DU Container and Send to WIPP**

This option consists of the following steps.

- *Isotopic dilution.* The  $^{233}\text{U}$  is mixed on a molecular scale with  $^{238}\text{U}$  and gadolinium—as described above. The product is a non-weapons-usable (<12 wt%  $^{233}\text{U}$  in  $^{238}\text{U}$ ), critically safe (gadolinium neutron absorber) solid. The oxide mixture is packaged into containers with up to 2 kg of  $^{233}\text{U}$  per container.

- *Packaging.* The solidified product is packaged in a strong container and placed in a 55-gal drum, which contains an internal DU metal or cermet liner. The DU liner must meet two functional requirements. First, there must be sufficient DU such as to provide sufficient radiation shielding to make the package a CH drum. Second, there must be sufficient DU such that if all the uranium in the drum (product package containing the down-blended  $^{233}\text{U}$  and the DU shielding liner) is combined, the  $^{233}\text{U}$  and  $^{235}\text{U}$  would be isotopically diluted with  $^{238}\text{U}$  such that nuclear criticality could never occur—independent of the gadolinium. The  $^{235}\text{U}$  is from (a) any  $^{235}\text{U}$  originally with the  $^{233}\text{U}$ , (b) the  $^{235}\text{U}$  in the DU that was blended with the  $^{233}\text{U}$ , and (c) the  $^{235}\text{U}$  in the DU shield.
- *Transport and disposal.* The final package is sent to WIPP as CH waste.

The  $^{233}\text{U}$  is made non-weapons usable by isotopic dilution. A fundamentally different criticality strategy is used with this option as compared to the previously discussed options. The gadolinium provides operational criticality control from the time the gadolinium is added in the process until long after the WIPP repository is closed. The DU shield provides added  $^{238}\text{U}$  for criticality control by isotopic dilution and thus helps ensure long-term geological criticality control—thousands to millions of years later. As the drum and its contents degrade over geological time, the  $^{233}\text{U}$ ,  $^{235}\text{U}$ , and  $^{238}\text{U}$  in the drum isotopically mix. Isotopic dilution as a method of criticality control provides the highest long-term margins against nuclear criticality since no mechanisms have been found in the natural environment that are capable of separating the  $^{233}\text{U}$  from the  $^{238}\text{U}$ .

The DU shield can be DU metal or a  $\text{DUO}_2$ -steel cermet (8). Cermets contain ceramic particulates ( $\text{DUO}_2$ ) embedded in a metal. The cermet manufacturing process results in a clean layer of steel on each side of the cermet and thus avoids potential contamination issues associated with the DU during handling operations. Cermets containing up to 90 vol %  $\text{UO}_2$  have been fabricated. Cermets provide an alternative method to incorporate DU into a ductile shielding material. Cermets have two advantages compared to uranium metal: (a) they are more chemically inert than uranium metal and (b) the uranium is in a chemical form that is more similar to that of the  $^{233}\text{U}$ . Uranium metal corrodes rapidly under certain anoxic conditions and thus there may be some constraints on its use in some applications. The use of a cermet avoids use and disposal issues associated with uranium metal (9).

There are significant shielding requirements. The occupational radiation issues are a consequence of two factors: (a) the  $^{232}\text{U}$  content of the  $^{233}\text{U}$  and (b) the placement of much larger quantities of  $^{233}\text{U}$  in a single drum. The radiation levels of  $^{233}\text{U}$  are significantly higher than those of Pu; however, if one were disposing of similar quantities of low-quality Pu in a single drum, occupational exposure would become a concern. Radiation levels are quantity- and impurity-level dependent.

Scoping calculations defined what such a package might look like if the product was from down-blending a single canister of CEUSP  $^{233}\text{U}$  (Table 1). The CEUSP material is the lowest-quality  $^{233}\text{U}$  in inventory and has some of the highest radiation levels. It is a complex mixture of  $^{233}\text{U}$ ,  $^{235}\text{U}$ , other uranium isotopes, gadolinium, and cadmium. A single canister contains 2.6 kg of uranium of which 0.25 kg is  $^{233}\text{U}$ . After processing the mixture to non-weapons-usable uranium and adding some gadolinium, a single CEUSP can yields 19.3 kg of product. One packaging option would be to place this product into a can 47.42 cm high and 9.94 cm in diameter, place the can in a 279.7 kg DU metal cylindrical shield (69.0 cm high and 32.5 cm in diameter), and place this assembly into a 208-l (55-gal) drum (84.8 cm high and 56.8 cm in diameter) with a drum weight of 30 kg. The drum would contain filler material between the DU shield and the inner drum wall. The total package weight is 329 kg. In this specific case, the quantity of DU needed for shielding is approximately equal to the quantity required for full isotopic dilution for long-term criticality control.

WIPP does allow shipment of wastes in larger containers than the standard 208-l drum. An example is the 10-Drum Overpack (TDOP). The option may exist to use a larger package with the isotopically diluted  $^{233}\text{U}$  in the center and the uranium shielding on the outside. By placing more  $^{233}\text{U}$  into a single larger package rather than several smaller packages, the same amount of shielding per unit of  $^{233}\text{U}$  will result in much lower external radiation doses. The definition of CH TRUW is a radiation dose of  $<200$  mrem/hr at contact. However, when multiple drums are stored or transported together, the total radiation levels from multiple drums may become much larger. If the drums are to be CH in this type of environment, the individual dose rate from any drum must be considerably  $<200$  mrem/r.

**Option 6: Isotopically Dilute  $^{233}\text{U}$  with  $^{238}\text{U}$  to Critically Safe, Non-Weapons-Usable  $^{233}\text{U}$ , Place in Drums, and Ship to WIPP**

This option consists of the following steps.

- *Isotopic dilution.* The  $^{233}\text{U}$  is mixed on a molecular scale with  $^{238}\text{U}$ —as described above except that it is blended with sufficient  $^{238}\text{U}$  such as to make the product critically safe by isotopic dilution with  $^{238}\text{U}$  ( $<0.66$  wt %  $^{233}\text{U}$  in  $^{238}\text{U}$ ) without the use of gadolinium. The typical assay will be  $\sim 0.5$  wt %  $^{233}\text{U}$  in DU.
- *Packaging and shipment.* The  $^{233}\text{U}$ - $^{238}\text{U}$  oxide is placed in WIPP disposal drums. The drums are shipped to WIPP

The  $^{233}\text{U}$  is non-weapons usable by isotopic dilution. The  $^{233}\text{U}$  is critically safe by isotopic dilution. The radiation doses will vary with time. At secular equilibrium, most of the drums will be RH drums. In the form of  $\text{UO}_3$ , the allowable  $^{232}\text{U}$  content in down-blended  $^{233}\text{U}$  that does not exceed the radiation limits for CH TRUW (200 mrem/h) for a fully loaded drum is between 0.01 and 0.02 ppm  $^{232}\text{U}$ . Before down-blending, this translates into 2 to 4 ppm  $^{232}\text{U}$  in the  $^{233}\text{U}$ . The  $^{232}\text{U}$  content of most of the inventory is higher than this value.

The production of a RH waste form has a major impact on drum storage, transport, and disposal. CH TRUW drums are transported using the TRUPACT II transport container. This container can accept 14 drums, and 3 such containers can be transported on a single truck. There are truck gross-weight limits that prohibit all of the drums being at the maximum drum weight limit. However, in practice, this is not usually a transport constraint because large sites ship a mixture of heavy and light TRUW drums in a single truck shipment. In contrast, the RH transport container can accept only three drums, and only one such container can be placed on a truck without exceeding weight limits. There are similar large differences in the handling impacts at WIPP of RH drums, as compared to CH drums. In addition, WIPP currently has excess capacity available for CH wastes but the inventories of RH wastes exceed currently available WIPP capacity.

## **Option 7: Isotopically Dilute $^{233}\text{U}$ with $^{238}\text{U}$ to Critically Safe, Non-Weapons-Usable $^{233}\text{U}$ , Mix with INEEL Sodium-Bearing Waste, Process and Package Mixture into Drums, and Ship to WIPP**

This option consists of the following steps.

- *Isotopic dilution.* The  $^{233}\text{U}$  is processed by one of several possible flow sheets at INEEL. The processing mixes  $^{233}\text{U}$  and  $^{238}\text{U}$ . The  $^{233}\text{U}$  is down-blended to be critically safe, non-weapons-usable  $^{233}\text{U}$  (<0.67 wt %  $^{233}\text{U}$  in  $^{238}\text{U}$ ). The most likely mixing process would be the full dissolution of the  $^{233}\text{U}$  and  $^{238}\text{U}$  in nitric acid.
- *Mixing with sodium-bearing waste.* The resultant homogeneous solution would be mixed with sodium-bearing liquid wastes at INEEL and solidified with those wastes.
- *Packaging and shipment.* The solidified product would be placed in WIPP disposal drums and shipped to WIPP.

The  $^{233}\text{U}$  is non-weapons usable by isotopic dilution. The  $^{233}\text{U}$  is made critically safe by isotopic dilution. The uranium mixture, after dissolution, is mixed with liquid-sodium-bearing wastes at INEEL. This is a large waste stream that may be classified as RH TRUW. The addition of the  $^{233}\text{U}$  stream to this existing waste stream may allow the process equipment and facilities that are used for the sodium-bearing waste to co-process the  $^{233}\text{U}$  waste with potential economic benefits. In some flow sheets, the uranium may simplify solidification of the sodium bearing wastes.

## **OBSERVATIONS AND CONCLUSIONS**

### **Viable Options**

Based on the limited available information, Option 5 (isotopically dilute  $^{233}\text{U}$  with  $^{238}\text{U}$  to non-weapons-usable  $^{233}\text{U}$ , place in shielded DU container, and send to WIPP) is likely to be the low-cost option:

- *Operations.* This option requires the least processing and handling of all the options that isotopically down-blend  $^{233}\text{U}$  to a non-weapons-usable material (<12 wt %  $^{233}\text{U}$  in  $^{238}\text{U}$ ). Processing is required. However, large-batch processing sizes can be used since the final product is not limited by WIPP criticality constraints to <200 g of  $^{233}\text{U}$  per container. There is no necessity to divide the product into small batches and separately package each small batch with <200 g of  $^{233}\text{U}$  equivalent—an expensive operation.
- *Disposal costs.* This option bypasses the drum criticality limits and thus minimizes the number of drums sent to WIPP.

The same option would likely minimize environmental, safety, and health (ES&H) concerns. The number of operations and potential exposures to workers is minimized. Transportation is minimized. Radiation exposure is minimized.

There are significant uncertainties associated with this option. Changes in WIPP WAC to allow isotopic dilution is required. The package is unusual. However, the changes that are required may provide higher margins of ES&H than other approaches.

## Institutional Issues

It is unclear that all the  $^{233}\text{U}$  can be considered defense TRUW suitable for disposal in WIPP under the current legal authorization. Many secondary wastes (HEPA filters, gloveboxes, etc.) from any processing option would clearly be high-alpha wastes ( $>>100$  nCi/g) but may not be TRUW. In this context, certain  $^{233}\text{U}$  wastes are orphan wastes. This is an institutional issue to be addressed by the Congress and the President.

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