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Science & Global Security: The Technical Basis for Arms Control, Disarmament, and Nonproliferation Initiatives

Publication details, including instructions for authors and subscription information: http://www.tandfonline.com/loi/gsgs20

Managing the Uranium-233 Stockpile of the United States

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To cite this article: Robert Alvarez (2013): Managing the Uranium-233 Stockpile of the United States, Science & Global Security: The Technical Basis for Arms Control, Disarmament, and Nonproliferation Initiatives, 21:1, 53-69

To link to this article: <u>http://dx.doi.org/10.1080/08929882.2013.754311</u>

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Managing the Uranium-233 Stockpile of the United States

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The United States produced about 2 tons of uranium-233, a weapons-useable fissile material, as part of its military and civilian nuclear program. Of that, 1.55 tons was separated at costs estimated to be between \$5.5 and \$11 billion. Of the 1.55 tons, approximately 96 kg of uranium-233 may be unaccounted for. There are also varying site-specific estimates suggesting that material control and accountability of the U.S. uranium-233 inventory needs to be more stringent. About 428 kg of uranium-233 is stored at the Oak Ridge National Laboratory (ORNL), in Tennessee at Building 3019, a 69-year-old structure which DOE describes as the "oldest operating nuclear facility in the World" and one that does not meet current safeguards and security requirements.

Currently, the U.S. Department of Energy's (DOE) goal for disposition of the 428 kg is 2018, more than 20 years after significant environmental, safety, and security vulnerabilities were first officially acknowledged. To meet this goal, DOE plans to waive its own waste acceptance criteria to allow direct shallow land disposal of a large portion of the uranium-233 by August 2014. Granting a disposal waiver sets a bad precedent for international safeguards and standards for the disposal of reprocessed wastes containing high concentrations of fissile materials.

INTRODUCTION

Uranium-233 was discovered in 1940 at the University of California at Berkeley and like plutonium-239 and uranium-235 it is fissionable and capable of fueling nuclear weapons or power reactors.

Uranium-233 is produced in a reactor or with an accelerator by bombarding thorium-232 with neutrons. Since thorium-232 is not fissile, it requires uranium-235 or other fissile materials to produce uranium-233. After a several-year cooling off period, uranium-233 can be recovered from the irradiated thorium through chemical separation in a nuclear reprocessing plant.

Received 5 August 2011; accepted 16 October 2012.

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Uranium-233 can be compared to plutonium for weapons-usability. As little as 0.5 kg of uranium-233 solution could initiate a nuclear criticality due to the presence of water. As metal, the minimum critical mass is about 6 kg¹. A 12 percent dilution with uranium-238 produces a critical mass similar to 20 percent enriched uranium, the upper safeguard boundary of low-enriched uranium.²

This article provides a brief overview of the history of production of uranium-233 by the United States and the estimates of the current inventory. It considers the history of U.S. efforts to develop uranium-233 as a power reactor fuel and assesses the extent to which safeguards and security concerns regarding the uranium-233 inventory have been addressed. The paper concludes with a discussion of the uranium-233 legacy and the shortcomings of DOE's current waste disposition plans

THE URANIUM-233 INVENTORY

From the 1940s until the mid to late 1950s, uranium-233 was produced in small quantities in the United States for weapons research and development (R&D). In 1955 the first uranium-233 nuclear device with a yield of 22 kilotons was detonated at the Nevada Atomic Proving Grounds.³ According to one U.S. nuclear weapons laboratory official, "uranium-233 has been shown to be highly satisfactory as a weapons material."⁴ By 1954, however, DOE's predecessor, the Atomic Energy Commission (AEC) had conducted a comprehensive study but did not pursue uranium-233 production for weapons use. A major factor was the radiation hazard from uranium-232 which is co-produced when irradiating thorium. "The gamma radiation associated with the uranium-232 chain" stated an expert at the AEC's Hanford site "is the major complication in the fabrication and utilization of uranium-233."⁵

In the early 1960s, there was renewed interest in using uranium-233 in nuclear weapons after "it was discovered that the plutonium components in the strategic warheads in stockpile were susceptible to serious damage when subjected to neutron irradiation induced fissions in space from nuclear-armed interceptor missiles in 'near-hit situations' within a ten mile radius." Because uranium-233 is more stable at high temperatures than plutonium, it raised the possibility that uranium-233 warheads would be more robust.⁶

This prompted the AEC nuclear production sites to develop techniques to produce "clean" uranium-233, with less than 5 ppm of uranium-232 to reduce radiation hazards. To achieve this goal, measures were proposed at the Hanford site, in which: (1) thorium obtained from pure monazites ore low in the thorium-230 isotope would be irradiated; (2) the material would be irradiated in neutron flux relative free of neutrons above 6 MeV, requiring a specially moderated reactor; and (3) irradiation of the thorium would be halted while it still held protactinium-233 (27-day half-life) that could be separated from uranium-232 before it decayed to uranium-233.⁷

By 1966, interest in large-scale production of uranium-233 for weapons use had waned but production for R&D continued. A review of the history of the DOE's Rocky Flats Plant, in Colorado, which manufactured weapons components, found that uranium-233 was being fabricated from 1965 until the early 1980s.⁸ From 1955 to 1968, several nuclear weapons tests were conducted using uranium-233.⁹

Uranium-233 was produced in more substantial amounts from 1965 to 1970 with the expectation that it could fuel power reactors. Developing uranium-233 as a potential fuel required the construction and operation of thorium fuel cycle R&D facilities, research reactors, radiochemical separations facilities, hot-cells, fuel fabrication facilities, and the storage of spent commercial reactor thorium fuel by DOE and its predecessor agencies.

Between 1954 and 1970 uranium-233 was produced in weapons material production reactors at the Savannah River Site (SRS) in South Carolina, the Hanford site in Washington, and in several commercial nuclear power plants (Indian Point I, Dresden I, Peach Bottom I, and Fort St. Vrain). Approximately two tons of uranium-233 was produced, of which 1556 kg was separated from 857 tons of thorium at reprocessing plants (Table 1), at an estimated total cost of \$5.5 to \$11 billion (2012 dollars) including associated cleanup costs.¹⁰ About 655 kg of separated uranium-233 subsequently fueled DOE research reactors and/or was lost to waste.¹¹ About 403 kg of uranium-233 was generated in commercial nuclear power and government reactors, but not reprocessed.¹²

As of 1999, approximately 805 kg of separated uranium 233 was stored at DOE sites.¹³ This leaves a discrepancy of approximately 96 kilograms or 6 percent of the total amount recovered from reprocessing plants. By comparison, the DOE's official inventory difference for plutonium published in 1996 is 2.1 percent.¹⁴

Uranium 233 as Power Reactor Fuel

In the 1960s and early 1970s the AEC assumed that 1000 GWe of nuclear capacity would be online inside the United States by the year 2000, with a similar nuclear capacity outside. As a result, the AEC predicted that world uranium supplies would be rapidly exhausted. These assumptions drove a two-track R&D effort for a new generation of reactors that would be capable of producing more fissile material than they consumed. As it turned out, the AEC's prediction of the year-2000 global nuclear capacity was off by an order of magnitude.

The first track was the development of fast-neutron reactors. "Fast" fission releases more neutrons per fission than the thermal neutron fission in

Site	Date (year)	Thorium Processed (tons)	Urainium-233 Recovered (kg)	Uranium-232 Content (ppm U)	Flowsheet Employed
Oak Ridge National Laboratory ¹	1954–1958	35	55	10–40	Interim 23 Thorex
Savannah River Plant ²	1964–1969	240	580	3.9–228	Interim 23 Thorex
Hanford Site ³	1965–1970	565	820	6–10	Interim 23 Acid Thorex
Nuclear Fuel Service (Indian Point I fuel) ⁴	1968–1969	17	101	125–144	Interim 23
		857	1556		

Table 1: Summary of Thorium–Uranium-233 Processing in the United States

Note. ¹Oak Ridge National Laboratory R.E. Brooksbank, W.T. McDuffee, R.H. Rainey, A Review of Thorium Fuel Reprocessing Experience, Oak Ridge TN, Conf-780223–3, January 1978.

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³Savanah River Plant, E.I. du Pont de Nemours and Company, D.A. Orth, *SRP Thorium Process-ing Experience*, Aiken, SC, June 1978.

⁴Oak Ridge National Laboratory, Chemical Technology Division Progress Report for the Period April 1983, to March 21, 1985, ORNL-6153, Oak Ridge, TN, October 1985.

conventional reactors. Plutonium-fueled fast-reactors in particular held the promise of producing electricity and 30 percent more fuel than they consumed.

The second track was the development of thorium based slow-neutron reactors. This led to the production of significant quantities of uranium-233 for reactor fuel. The attractiveness of the thorium fuel cycle was due to the abundant supply of thorium in nature and the potential of uranium-233 to increase the efficiency of fissile material production in slow-neutron reactors relative to uranium-235 and plutonium. For example, although current light water reactors generate plutonium-239, only about 0.6 atoms are produced per fission of uranium-235 or plutonium-239 (a conversion ratio of ~0.6). For thorium the conversion ratio can be greater than one, holding the promise that such a reactor can produce more fissile material than it destroys.

R&D of several reactor types was launched with the goal of demonstrating that uranium-233 derived from thorium would be a safe and economical source of electricity. Projects demonstrating the potential viability of slowneutron breeder reactors using uranium-233 were established, most notably the Elk River Reactor in Minnesota, the Molten Salt Reactor at ORNL, and the Light Water Breeder Reactor at Shippingport, Pennsylvania.¹⁵ By 1977, however, pursuit of the thorium fuel cycle was effectively abandoned in favor of plutonium-fueled fast-reactors. All told the AEC and its successor the DOE spent billions of dollars on thorium fuel cycle R&D.

Another factor that may have influenced the decision to abandon the thorium fuel cycle is that thorium is more radioactive than uranium and requires additional safeguards. The surface dose rate from a 55 gallon drum of thorium oxide is approximately 60 mR/hr, about 13 times higher than a similar sized drum of uranium.¹⁶ A worker spending time inside a thorium storage facility could expect to encounter dose rates of 60–100 mR/hr, reaching the U.S. occupational exposure limit of 5 rem in just over 6 days.

The Peach Bottom I Unit, a prototype 40 megawatt high-temperature gascooled reactor used thorium fuel. It operated from 1967 to 1974. The reactor was closed after experiencing a high rate of fuel element failures, causing significant down-time.²² Its spent fuel is currently stored at DOE's Idaho National Engineering Laboratory (INEL).²³

The Fort St. Vrain plant, operating from 1979 to 1989, was a hightemperature-gas-cooled 330 megawatt reactor using thorium and uranium-235 fuels. Hundreds of events involving equipment failure, gas leaks, fuel failures, cracked piping and graphite, and human error led to its closure.²⁴ DOE is responsible for its spent fuel which is stored in dry casks at the reactor site.²⁵

In June 2000, after lobbying by ORNL, the Secretary of Energy directed the Office of Nuclear Energy to utilize the stockpile of excess uranium-233 to increase the supply of medical isotopes. This was a multi-step process in different locations requiring new facilities to extract thorium-229, an 8,000-year half-life decay product of 160,000-year half-life uranium-233, which then decays into actinium-225 (10-day half-life), and subsequently decays, through short-lived intermediates, into bismuth-213 (46 minute half-life), a radioiso-tope of potential interest for medical therapy.²⁶ In January 2001, the DOE determined, however, that other than medical purposes, there was no programmatic use for the uranium-233 stored at ORNL.²⁷ In 2005, the U.S. Congress

terminated medical isotope production from the ORNL inventory and transferred the responsibility for its disposition to the Office of Environmental Management (EM).²⁸ That year, Congress also directed the U.S. Defense Department to dispose of 3,222 tons of thorium in the U.S. strategic material stockpile.²⁹

SAFEGUARDS AND SECURITY

Separated uranium-233 is classified by the U.S. government as a Category I strategic special nuclear material, i.e., material that "in specified forms and quantities, can be used to construct an improvised nuclear device capable of producing a nuclear explosion."³⁰ The United States and the International Atomic Energy Agency (IAEA) require safeguards for uranium-233 in quantities greater than 2 Kg.³¹ A Category I quantity of highly enriched uranium (HEU) is 5 kg. According to DOE, stringent physical security is required for uranium-233 stocks, in order to prevent "an unauthorized opportunity to initiate or credibly threaten to initiate a nuclear dispersal or detonation ... for onsite assembly of an improvised nuclear device."³²

Interest waned for weapons use of uranium-233 due to it radiological hazards, and the related costs of safeguarding the materials and protecting worker safety. Of particular concern is exposure to uranium-232, which is co-produced and is 60 million times more radioactive than uranium-238. This is due to the short half-life of U-232 and the high-energy gamma radiation emitted in the decay scheme of uranium-232 daughter products, most notably thallium-208, which emits a 2.6 MeV gamma ray when it decays. This gamma radiation is difficult to shield. With a half-life of 72 years, the decay of uranium-232 increases external penetrating (gamma) radiation levels, which peak after 10 years (Figure 1).

The CEUSP material in Building 3019 was packaged over 25 years ago and contains the highest concentration of fissile material (uranium-233/235) in the facility—approximately 86 percent of the total uranium content.

The uranium-232 contaminant level, however, is not considered an adequate barrier to prevent a terrorist from making an improvised nuclear device. According to researchers at ORNL, "... if a diverter was motivated by foreign nationalistic purposes, personnel exposure would be of no concerns since exposure even that these levels would not result in immediate death."³³ Uranium-233 currently stored at DOE sites typically contains from 6 to 162 parts per million of uranium-232, making it vulnerable to seizure by terrorists.³⁴ This is underscored by the DOE's experience with handling uranium-232 contaminant levels to make weapons components that were comparable to those being stored in Building 3019 at ORNL. For instance, during the 1960s and 1970s the DOE's Rocky Flats Plant processed uranium-233 in uranyl nitrate and then

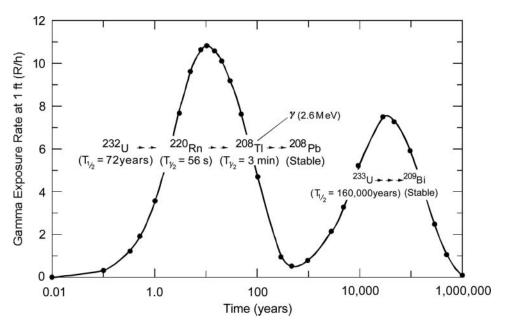


Figure 1: Gamma exposure rate for uranium-233 (ORNL/TM-13553).

fabricated uranium-233 metal components in kilogram quantities. According to a DOE report about handling uranium-233 at Rocky Flats:

The material also contained approximately 50 parts per million (ppm) contaminant.... A 50 ppm U-232 content equates to approximately 13 R/hr at 1 foot and with extrapolation, a 5 to 10 ppm content would emit approximately 5 R/hr [at 1 foot].³⁵

Under this circumstance, it would take about one to two weeks of exposure, working 12 hours-a-day, before a person would accumulate a potentially lethal dose.

Managing the Uranium-233 Legacy

The lack of an accurate uranium-233 inventory has raised concerns. In 1996, the U.S. DOE's Inspector General (IG) issued an audit report that concluded: "management at three sites [ORNL, Rocky Flats and the Idaho National Laboratory] had not performed all required physical inventories, and one site did not perform measurements, due to safety concerns and operational interruptions ... the longer complete physical inventories are delayed, the greater the risk that unauthorized movement of special nuclear materials could occur and go undetected."³⁶

In the years following the IG's audit, DOE has reported varying estimates of the inventory at Oak Ridge and Idaho. For instance, published sources for uranium-233 stored in Building 3019 at ORNL range from 359 kg³⁷ to 450 kg,³⁸ an uncertainty of 91 kg. At DOE's Idaho National Laboratory, inventory data of un-irradiated uranium-233 range from 352 kg³⁹ to 411.36 kg⁴⁰, an uncertainty of 59.36 kg.

It is possible that more uranium-233 was lost to waste than publically reported or is in diluted forms. Also the quantity at the Los Alamos National Laboratory (LANL) in 1997 and the amount of uranium-233 used in weapons tests remains classified. According to the DOE, prior to 1970, when all uranium-233 was produced, material measurement technologies "were less accurate than today."⁴¹

There are 1,100 containers with uranium-233 stored in Building 3019.⁴² It is also eligible for listing in the National Register of Historic Places.⁴³ DOE finds that this facility has "deteriorated beyond cost-effective repair. Significant annual costs would be incurred to satisfy current DOE storage standards, and to provide continued protection against potential nuclear criticality accidents or theft of the material."

Surface dose rates from the Building 3019 canisters range between 1 to 300 R per hour.⁴⁴ About 500 canisters are holding what is considered "high purity" material (~197 kg) that contains less than 50 ppm uranium-232.⁴⁵ Building 3019, which holds approximately 1,000 kilograms of uranium-233 and uranium-235 does not meet current Category I security requirements. For example, a batch of material that came from the Indian Point I reactor is co-mingled with nearly eight times more uranium-235 (796 kg) in HEU, another Category I fissile material.⁴⁶ Nor does it meet physical security requirements of the IAEA, which includes intrusion surveillance and motion detection.⁴⁷

In 1976, after the DOE abandoned development of the thorium nuclear fuel cycle, efforts were made to stabilize some of the excess uranium-233 at ORNL. Other than that, the US stockpile of excess uranium-233 was subject to decades of neglect. In December 1996, the DOE's Office of Environment, Safety and Health conducted a vulnerability assessment of the storage of HEU at its sites. It found that inventories of separated uranium-233 at several sites were being stored in conditions that increased environmental, safety, and health risks. According to the assessment:

Some of the significant ES&H vulnerabilities involve uranium-233.... Fourteen vulnerabilities involving uranium-233 were identified. Four of these are among the most significant found in the assessment. Uranium-233 has been stored in metal containers, outside on pads, buried in drums in earthen mounds, or stored for decades without inspection.⁴⁸

Of particular concern were the inventories at ORNL Building 3019, constructed in 1943, and holding 1,100 "cans" of uranium-233 (Figure 2). This

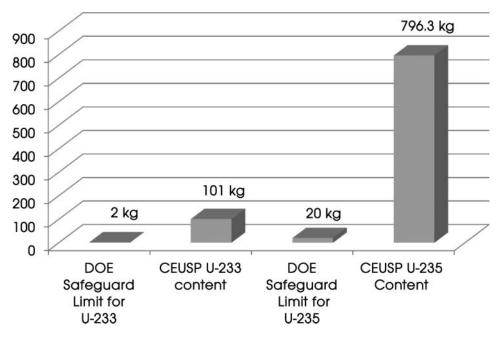


Figure 2: Landfill disposal of CEUSP material exceeds DOE Safeguard Termination Limits.

facility was originally designed as a radiochemical processing facility and not a long-term storage facility for approximately 1,000 kg (uranium-233/235) Category I fissile materials. In the early 1990s it was designated as DOE's "national repository for uranium-233" and contains material from SRS, ORNL, Rocky Flats, Lawrence Livermore National Laboratory, and LANL in the form of oxides and metals.

Three years earlier in 1993, a DOE review of Building 3019 found the safety documentation to be "adequate."⁴⁹ The 1996 DOE vulnerability assessment found that despite the facility's nuclear safety paper-work, an environmental release from the containers "could be expected to occur within the next five years in that some of the packages are approaching 30 years of age and have not been regularly inspected." ⁵⁰

Concurrently, the DOE's Defense Nuclear Facilities Safety Board (DNFSB) issued a critical report about the safety of the DOE's uranium-233 inventory highlighting the problems in Building 3019.⁵¹ As a result, in 1997 the Board issued a recommendation that urged the DOE to establish an agency-wide project to correct storage vulnerabilities.⁵² Building 3019 at ONRL had among the most significant vulnerabilities.

The risks at Building 3019 include natural events (earthquakes, tornados) fires, explosions and nuclear criticalities. Consequences from possible fires, explosions, and other container failures holding uranium-233/235 recovered from Indian Point I reactor fuel (CEUSP material) are estimated to result in

potential offsite doses of 45 rems at a distance of nearly 6 miles,⁵³ 450 times the annual public exposure limit at DOE sites.⁵⁴ It took eleven years before the DNFSB closed its recommendation.⁵⁵ Yet the conditions of the building have deteriorated, while contractor management and costs continue to escalate.⁵⁶

A major reason the project is considered low priority is that DOE selfregulates and considers the effort to process and dispose of this material an "unfunded mandate" not linked to established program activities including environmental compliance agreements. Although the DNFSB can issue recommendations it does not have regulatory authority to establish deadlines or issue fines and penalties.

In November 2005, after fits and starts, EM assumed control of the project to downblend and dispose of the uranium-233. Nearly five years later, DOE's IG reported that, "the Department's uranium-233 disposition project had encountered a number of design delays, may exceed original cost estimates, and will likely not meet completion milestones." Since EM took over this project, it was directed by four DOE managers in less than two years and the estimated total cost of the project increased from \$384 million to \$473 million.⁵⁷

In 2009, in an effort to reduce costs, DOE developed a plan for uranium-233 disposition 58 in which:

- CEUSP material (403 canisters and 73 percent of the fissile inventory in Building 3019) would be directly disposed in a landfill at the Nevada National Security Site (NNSS).⁵⁹
- The DOE's National Nuclear Security Agency would assume responsibility for approximately 245 kg of uranium-233 oxide contained in Zero Power Reactor fuel plates left over from the Molten Salt Reactor Experiment and transfer it to the high-security Device Assembly Facility (DAF) at NNSS.
- The remaining uranium-233 with uranium-238 oxides would be downblended using existing hot cells at ORNL or with aqueous downblending followed by co-processing with radioactive waste sludges stored at the ORNL site.

In January 2012, DOE announced it had shipped six canisters containing the Zero Power Reactor plates to the DAF at the NSSS for use in experiments.⁶⁰ Shipment of the remaining 122 plates containing uranium-233 were completed by the end of June 2012.⁶¹ The goal for final removal and disposition of the balance of material in Building 3019 is projected for 2018.⁶²

DOE appears set on the direct disposal option for the CEUSP material in a land fill, even though it would significantly violate its own safeguard and security requirements⁶³ and NNSS Waste Acceptance Criteria (WAC)⁶⁴ (Figures 2 and 3). DOE argues that the CEUSP material would be placed in slit trenches, up to 40 feet deep and then covered with low-level radioactive waste

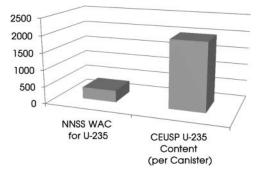


Figure 3: Disposal of CEUSP material exceeds DOE's Waste Acceptance Criteria for landfill disposal of fissile materials.

to serve as an additional barrier to prevent intrusion.⁶⁵ However, uranium-233 and uranium-235 have half-lives of 160,000 years and 7044 million years respectively. The National Research Council concluded in 2001 that "DOE's preferred solutions—reliance on engineered barrier and institutional controls are inherently failure prone."⁶⁶ The bigger issue may be that weapons-usable uranium-235, which serves as a diluent for the uranium-233, is going into a land disposal site without security.⁶⁷ DOE is currently reviewing the potential for an intruder obtaining the CEUSP material after it is disposed.⁶⁸

The contents in the CEUSP canisters are Category I material and meet the criteria for "high-grade material." For high-grade material, DOE requires safeguards for quantities of uranium-233 greater than 2 kg and quantities of uranium-235 greater than 20 kg. In order to dispose of this material, the DOE will have to grant an unprecedented termination of safeguard requirements.

The uranium-233 inventory of 323 kg at INEL was diluted in 31 tons of unirradiated thorium (1 percent dilution) and shipped to the NNSS beginning in 2010, where it was disposed as low-level radioactive wastes.^{69,70}

According to the NNSS WAC, uranium-235 disposal is limited to no more than 350 grams per package. The CEUSP material in Building 3018 at has an average of 2,000 grams of uranium-235 and about 250 grams of uranium-233 per package. The NNSS WAC is silent about uranium-233 but uranium-233 has a critical mass much less than uranium-235. The DOE has not revealed how it will reconcile this. Meanwhile, over the past fifteen years DOE has spent approximately \$84 million on the surveillance of the uranium-233 packages at ORNL.⁷¹ These costs are expected to increase as Building 3019 deteriorates. Since the CEUSP material is highly radioactive and was reprocessed from spent fuel, it could arguably be considered high-level radioactive waste (requiring geologic disposal), as defined in the Nuclear Waste Policy Act of 1982.⁷² Other than the fissile material in ORNL's Building 3019, DOE has pursued a policy of ultimate geologic disposal for excess weapons-usable fissile materials by blending down HEU and developing mixed oxide fuel or

direct disposal of diluted plutonium in the Waste Isolation Pilot Project (WIPP) a geologic disposal site in New Mexico. In 1998 the DOE approved termination of safeguards for residues bearing plutonium for disposal at the WIPP. Termination was granted under the condition that the residues would be downblended to "below 10 weight percent plutonium and placing the blended residues in the pipe overpack containers prior to removing the residues from the protected area."⁷³

DOE plans to complete shipment of the CEUSP material for land-fill disposal by August 2014.⁷⁴ If a variance terminating safeguards for the CEUSP material is implemented, its fissile concentration would be more than eight times greater than allowed for the Rocky Flats material. Moreover the radiation barrier created by contamination from uranium-232 will diminish by 50 percent in less than 50 years, allowing for much easier access to the material for use in weapons.

CONCLUSION

The storage of the U.S. stockpile of uranium-233 is a safeguard, security, and safety risk and the production of the stockpile has left a disposal burden. The key concerns are: uranium-233 inventory problems (with possibly 123 kg unaccounted for in the DOE complex); inadequate protection of weapons-grade material; environmental, safety and health risks; the liability for management and disposal of spent thorium fuel from several commercial reactors; and disposal of uranium-233, including in landfill. Put simply, to save a few bucks the United States has decided no guns, gates, or guards for a lot of bombs worth of fissile material.⁷⁵

Nuclear material control and accountancy of uranium-233 is a key element in assuring the adequate safeguard and security of this fissile material. The DOE should provide a more accurate inventory. Disposal of large quantities of concentrated uranium-233 waste in a land fill would significantly violate the DOE's safeguard and security requirements and NNNS Site WAC. In order to dispose of this material the DOE will have to grant an unprecedented termination of safeguard requirements, which would be unwise since it would set a bad precedent for safeguarding and disposal of other wastes containing significant amounts of fissile materials. A policy that allows for the permanent shallow land disposal of 897.3 kilograms of concentrated nuclear explosive material places the United States in an impossible position to criticize the nuclear materials security of other countries.

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