

THORIUM NITRATE STOCKPILE—FROM HERE TO ETERNITY

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ABSTRACT

The ^{232}Th nuclide is naturally occurring, with a half-life of 1.40×10^{10} years. The thorium series has ^{232}Th as the parent and, after a series of alpha and beta emissions, reaches ^{208}Pb , a stable end product. In many steps in the series, gamma rays and conversion electrons are emitted.

The Defense National Stockpile Center (DNSC), a field level activity of the Defense Logistics Agency (DLA) has stewardship of a stockpile of thorium nitrate that has been in storage for decades. The stockpile is made up of approximately 3.2 million kg (7 million lb) of thorium nitrate crystals (hydrate form) stored at two depot locations in the United States. The material is stored in several configurations in over 21,000 drums. The gamma dose at the surface of an isolated drum is 0.2–0.3 mSv/h (20–30 mR/h); however, the gamma dose within the storage facilities is 0.6–1.0 mSv/h (60–100 mR/h). The stockpile is classified as source material (10 CFR 20.1003) and is regulated by the U.S. Nuclear Regulatory Commission. The entire stockpile has been declared surplus to the needs of the Department of Defense.

DNSC sought technical assistance from Oak Ridge National Laboratory (ORNL) to define and quantify the management options for the thorium nitrate stockpile. The project objectives included: (1) developing the technical base to allow the DNSC to make informed decisions; (2) identifying potential disposal and storage options; and (3) establishing methods to determine the status of the stored material, from characterization through acceptance as a waste form at a disposal site. The program was broken into three phases: (1) historical data assembly and structured option analysis, (2) stockpile characterization, and (3) stockpile disposition. Phases 1 and 2 are complete. Analyses in Phase 1 demonstrated that disposition of the thorium nitrate inventory as a containerized waste, without processing, is the least complex and lowest-cost disposal option. Assessing the viability of this and other disposition pathways required a determination of which portion, if any, of the thorium nitrate inventory exhibits hazardous characteristics.

This paper describes methodologies and results comprising the work in Phases 1 and 2. The results allow the DNSC to structure and schedule needed tasks to ensure continued safe long-term storage and/or phased disposal of the stockpile.

The assistance provided by ORNL helped to solve several problems by (1) collecting and packaging existing information about the physical forms and storage characteristics of a 40+ year old inventory; (2) defining pathways, including endpoints, for safe long-term storage and/or disposition; (3) establishing a sampling and analysis program to define physical forms, radiological constituents, and potentially hazardous characteristics; (4) developing flowsheets for chemical processing to remove hazardous characteristics; and (5) merging historical information with newly performed calculations and newly acquired data to demonstrate compliance with waste acceptance criteria.

INTRODUCTION

The Defense Logistics Agency (DLA) Defense National Stockpile Center (DNSC) has stewardship of a stockpile of thorium nitrate that has been in storage for decades. The stockpile consists of approximately 3.2 million kg (7 million lb) of thorium nitrate crystals (hydrate form) stored in over 21,000 drums in several configurations (see Fig. 1) at two depot locations in the United States: Curtis Bay, Maryland, (75% by weight of the stockpile) and Hammond, Indiana (25% by weight). The gamma dose at the surface of an isolated drum is 0.2–0.3 mSv/h (20–30 mR/h); however, the gamma dose within the storage facilities is (0.6–1.0 mSv/h) 60–100 mR/h.

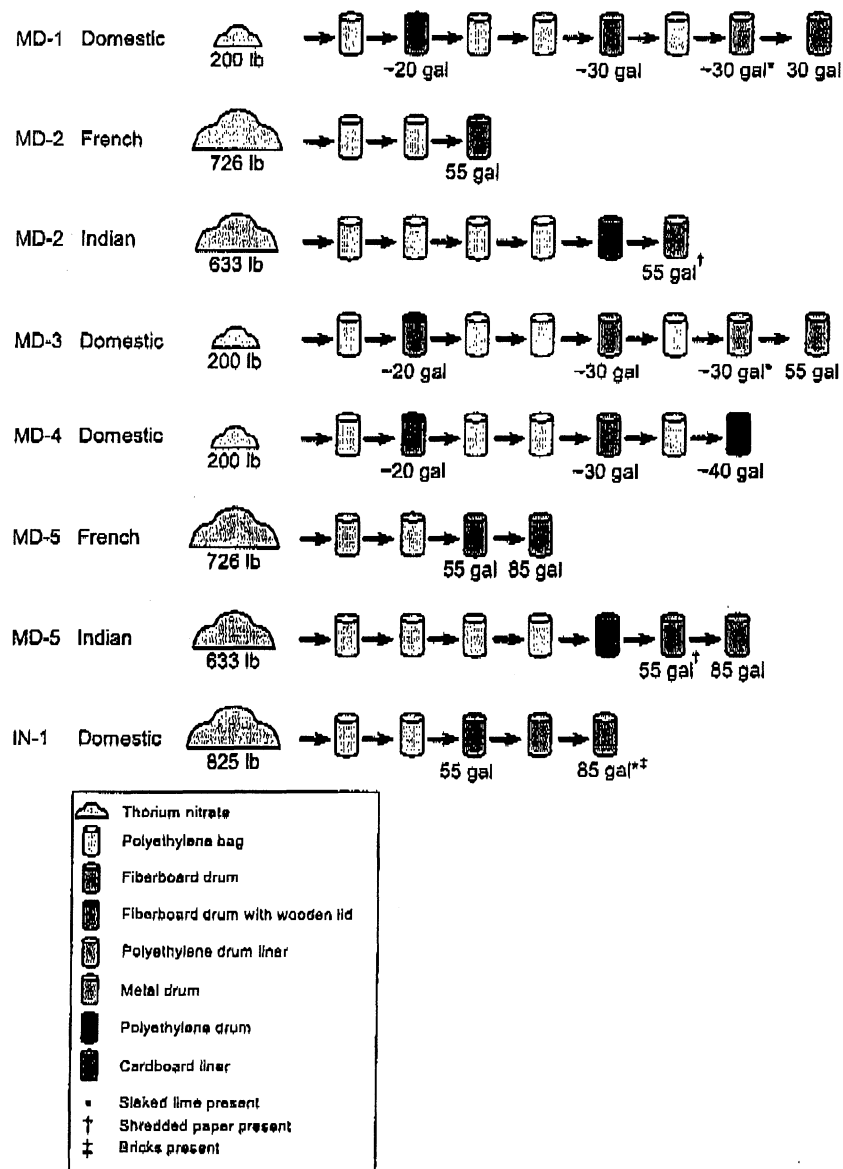


Fig. 1. Drum package configurations used in the storage of the thorium nitrate stockpile.

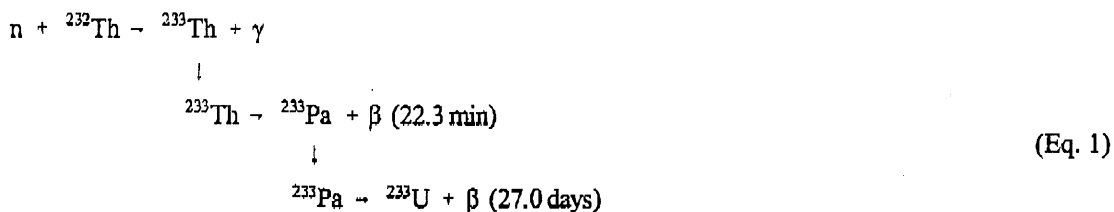
The thorium nitrate stockpile was produced from 1959 to 1964 for the Atomic Energy Commission (AEC) and previously has been under the control of several federal agencies. The entire stockpile—which is defined as source material (10 CFR 20.1003) and regulated by the U.S. Nuclear Regulatory Commission—has been declared by Congress to be surplus to the needs of the Department of Defense (DoD). Oak Ridge National Laboratory (ORNL) has been assisting DNSC by performing a three-phase project to analyze the bounds of possible and appropriate long-term management options for the thorium nitrate stockpile. The DNSC has historically sold excess thorium nitrate to domestic and foreign companies, but there is no current demand for this material.

This paper presents background information and summarizes the findings of the ORNL studies regarding the stockpile, including:

- the ^{232}Th decay chain and nuclear implications;
- DNSC stockpile management;
- disposition analysis program;
- Phase 1—historical data assembly, process characterization, and structured option analysis;
- Phase 2—stockpile characterization;
- modeling for disposal; and
- conclusions.

THORIUM-232 DECAY CHAIN AND NUCLEAR IMPLICATIONS

The ^{232}Th nuclide is naturally occurring, with a half-life of 1.40×10^{10} years. The thorium series has ^{232}Th as the parent and, after a series of alpha and beta emissions, reaches ^{208}Pb , a stable end product. In many steps in the series, gamma rays and conversion electrons are emitted. The ^{232}Th is not a fissile isotope but can be used as a precursor to generate fissionable ^{233}U , which can sustain a chain reaction in a nuclear reactor. Uranium-233 generates significantly more neutrons during thermal fission than either ^{235}U or ^{239}Pu , making it a desirable fuel to sustain criticality, while having excess neutrons to breed more ^{233}U from thorium. The reaction sequence to transform the fertile ^{232}Th into the fissionable ^{233}U is as follows:



When irradiated with neutrons, the ^{232}Th nucleus captures a neutron to become ^{233}Th . One-half of the ^{233}Th decays every 22.3 min by emitting an electron and transforming into ^{233}Pa . The newly formed ^{233}Pa decays with a half-life of 27 days by emitting an electron to generate the fissile ^{233}U .

As shown in the following decay chains, a small fraction of the ^{232}Th will capture a neutron followed by the emission of two neutrons. The resulting ^{231}Th will decay to the long-lived ^{231}Pa (half-life of 3.276×10^4 years) and accumulate. The ^{231}Pa has a significant cross-section for neutron capture (210 barns) and over time will transform in the reactor to ^{232}Pa , which will, in turn, decay to ^{232}U by emission of a beta particle.

One progeny in the decay chain is ^{220}Rn , which, as a gas, can escape and easily spread contamination. The decay chain continues beyond the ^{220}Rn gas and finally generates ^{208}Tl , which emits a highly penetrating 2.6-MeV gamma ray. Because the ^{232}U concentration increases gradually throughout the fuel burn-up, the handling of thorium spent nuclear fuel or the recovery of ^{233}U has to address both the ^{220}Rn containment as well as the high radiation doses from its progeny ^{208}Tl . This issue is both a major problem—because of the potential contamination and exposure risks to personnel and environment—and a potentially advantageous factor—because it greatly diminishes the value of using $^{233}\text{U}/^{232}\text{U}$ in nuclear weapons (i.e., it serves as a proliferation deterrent).

DNSC—STOCKPILE MANAGEMENT

The DNSC sought technical assistance from ORNL to quantify the thorium nitrate stockpile management options, including (1) developing the technical base to allow the DNSC to make informed decisions; (2) determining potential options for long-term management, including disposal and/or storage; and (3) establishing methods to determine the status of the stored material, from characterization through acceptance as a waste form at a disposal site.

The program was divided into three phases: (1) Historical Data Assembly, Process Characterization, and Structured Option Analysis; (2) Stockpile Characterization; and (3) Stockpile Disposition. Phase 1 is complete and Phase 2 will be completed in fiscal year (FY) 2003, and the DNSC plans to utilize the results of those assessments to make programmatic decisions and structure Phase 3, Stockpile Disposition.

DISPOSITION ANALYSIS PROGRAM

The disposition analysis program at ORNL defined and characterized potential disposition pathways for the thorium nitrate stockpile. During that process, ORNL identified and solved several problems: (1) determining existing information for 40+ year old inventory by collecting and documenting existing information from physical forms and packaging; (2) defining pathways, including endpoints, for safe long-term storage and/or disposal; (3) developing a sampling and analysis program to define physical forms, radiological constituents, and potentially hazardous characteristics; (4) preparing flowsheets for chemical processing to remove hazardous characteristics; (5) establishing methods for safely handling and depressurizing pressurized drums, (6) merging historical information with newly performed calculations and newly acquired data to demonstrate compliance with waste acceptance criteria (WAC), and (7) identifying transport requirements and specific packaging concepts for future updates to meet Department of Transportation (DOT) requirements.

PHASE 1—HISTORICAL DATA ASSEMBLY, PROCESS CHARACTERIZATION, AND STRUCTURED OPTION ANALYSIS

During Phase 1 of the program, the stockpile origination history was investigated as part of ORNL's historical data assembly activities to establish the AEC tie and time line. The information summarized in Table I clearly documents that the thorium nitrate stockpile was produced for the AEC.

In related Phase 1 tasks, ORNL identified over 30 potential disposition pathways for the stockpile. These pathways were consolidated into four options: dispose as thorium nitrate, long-term storage as thorium nitrate, process to a more stable chemical form for disposal, and process to a more stable chemical form for long-term storage.

Table I. Contracts on U.S. Government Procurement of Thorium Nitrate^a

Purchase contract number and country of origin	Thorium nitrate mass, kg (lb)	Information source/comment(s)
<i>Material stored at Curtis Bay</i>		
BSD-AE-57-47 United States	1,850,441 (4,079,456) in 20,400 drums	Notebook Sect. 4.4, Brussolo and Moore memo (5/17/60) and Notebook Sect. 6.1, Van Bergen memo (5/9/60). Compares well with maximum historical mass inventory reported in Table 3.1b of thorium nitrate inventory definition report (IDR). ^b This mass is about 2,950 kg (6,500 lb) less than what is reported in Table 3 of Taesin Chung's article. ^c This material was produced by the Lindsay Chemical Co. at the West Chicago Facility for the U.S. Atomic Energy Commission (AEC). Between 1959 and 1962, this material was delivered from West Chicago to the National Defense Stockpile. It was initially accepted by the AEC New Brunswick Laboratory for analysis and later transferred to Curtis Bay.
BSD-SM-61-18 India	218,862 (482,502) in 761 drums	Notebook Sect. 1.2, second table. Agrees with maximum Indian inventory in Table 3.1d of IDR. This material was purchased by the Commodity Credit Corp. (a U.S. government purchaser) through India's State Trading Corp. and delivered to the National Defense Stockpile in 1962. The material was initially accepted by the AEC New Brunswick Laboratory for analysis and later transferred to Curtis Bay.
BSD-SM-62-12 France	915,706 (2,018,753) in 2,776 drums	Notebook Sect. 1.2, third table. Barter specification BA-SP-1a (dated 2-10-61). Agrees with maximum French inventory in Table 3.1c of IDR. This material was purchased by the International Selling Corp. (New York, New York) through the French agency Commissariat a l' Energy Atomique and delivered to the National Defense Stockpile during 1962-1963. Some of the inventory was sampled at the AEC New Brunswick Laboratory.
<i>Material stored at Hammond</i>		
BSD-SM-62-14 United States	925,083 (2,039,400) in 2,472 drums	Notebook Sects. 3.1 and 3.2. (Hammond Inventory) From table in Sect. 3.1, 50 lots × 18,337 kg/lot + 8,233 kg = 925,083 kg (50 lots × 40,425 lb/lot + 18,150 lb = 2,039,400 lb). Agrees with maximum Hammond inventory reported in Table 4.1a of IDR and with Table 3 of Taesin Chung's article. This material was produced by the American Potash and Chemical Corporation at the West Chicago Facility for the U.S. AEC. Between 1962 and 1964, this material was delivered from West Chicago to the National Defense Stockpile. Some of this inventory was sampled at the AEC New Brunswick Laboratory.

^aBased on information contained in a notebook of historical DLA/DNSC records, "Historical Records of the DLA/DNSC Thorium Nitrate Inventory."

^bW. H. Hermes, et al. *Thorium Nitrate Material Inventory Definition Report*, ORNL/TM-2000/163, Oak Ridge National Laboratory, Oak Ridge, Tenn., June 2001.

^cT. Chung. "The Role of Thorium in Nuclear Energy," pp. ix-xvii, in *Uranium Industry Annual 1996*, DOE/EIA-0478(96), U.S. Department of Energy, Washington, D.C. April 1997. (Note. This report documents the domestic inventories from the West Chicago Facility, but does not track material received from outside the U.S.)

Preliminary design elements, including processing flowsheets, were developed for the four options, and estimated life-cycle costs were prepared for each option. The options were rated based upon waste quantity, health and safety impacts, environmental impacts, program risks and impacts, and permitting risk. Figure 2 displays the results of the ratings.

Option	Relative Cost	Waste Quantity (ft) LLW/ Hazardous/ Mixed	Health and Safety Impacts	Environmental Impacts	Program Risks and Impacts	Permitting Risk
A1 - ThN as LLW to NTS/dispose		328,800/ 0/ 0				
A2 - ThN as LLW to commercial disposal site/dispose		438,000/ 0/ 0				
A3 - ThN as LLW to uranium mill/dispose		114,000/ 0/ 0				
B1 - Baseline - continued storage at depots/store		NA*				
B2 - ThN to new off-site commercial facility/store		NA*				
B3 - ThN to off-site Govn-owned facility/store		NA*				
C1 - ThN as LLW - process for disposal at uranium mill/dispose		60,000/ 55,000/ 21,000				
C2 - ThN as mixed waste - process for disposal at NTS/dispose		60,000/ 55,000/ 21,000				
C3 - ThN as mixed waste - process at commercial disposal site/dispose		337,000/ 55,000/ 0				
D1 - ThN as source material - commercial process for safe bulk storage/store		21,000/ 55,000/ 0				

*Minor volume of PPE from repackaging

Key: Major Improvement Some Improvement Change Some Decline Major Decline

ORNL DWG 2001-48A
NA - Not applicable

Fig. 2. Life-cycle option ratings summary.

A favorable option has been identified by ORNL based on life-cycle analysis methods (see Fig. 2 for comparative results). This option can be described as whole drum burial in an arid environment, far removed from the water table, and in a remote location. The radiological hazards have been defined in ORNL reports,* with the key finding that disposal methods are favored if those methods can obviate the need for repackaging and/or opening drums and pre-treating the material. Chemical stabilization and/or conversion for disposal would involve increased technical and human risks, and 5-10 times the cost compared with as-drummed disposal in the thorium nitrate fused crystallized form. The direct as-drummed technical approach for disposal simplifies handling, reduces cost, and minimizes risk for worker alpha uptake and/or gamma dose. The whole drum disposal option includes characterization, packaging, and transport. This finding is supported by the preliminary chemical and radiological characterization data established by a sampling and analysis campaign.

The characterization work scope completed in 2002 included sampling over 100 drums for Resource Conservation and Recovery Act (RCRA) chemical analyses and for radiological characterization. Salt block-like monoliths comprise the predominant physical form, and a significant number of the Curtis Bay 30-gal drums are pressurized. Special container inspection and dose management assessments were completed to meet on-site material handling, transportation, and disposal site requirements. The Nevada Test Site (NTS) was utilized as a case study for the disposal site. Special radiological analysis, waste acceptance, and certification requirements are defined for potential disposal program implementation.

A disposal facility operated by the U.S. Department of Energy (DOE)/National Nuclear Security Administration at NTS could accept thorium nitrate for disposal, with the agreement of DoD and DOE. Such an agreement may be feasible because the thorium nitrate material was originally processed for the AEC, a predecessor of DOE. Thorium nitrate meets the NTS WAC for low-level radioactive waste, and the waste form is acceptable for disposal with the current physical, chemical, and radiological characteristics. Inspection, packaging, and transportation requirements for disposal of the thorium nitrate have been identified, and feasible methods are available and economically reasonable. Additionally, NTS provides a disposal location that is remote, well characterized, hundreds of feet from groundwater, and controlled. Bulk disposal of the as-drummed thorium nitrate drums is reasonable for the entire inventory.

PHASE 2—STOCKPILE CHARACTERIZATION

After Phase 1 was completed, it was apparent that the existing characterization of the stockpile was insufficient to support decisions regarding its disposal. Progress in analytical chemistry since the material was produced would allow the production of more accurate data; therefore, the main objective for FY 2002 was to develop and implement a sampling and analysis plan that would yield objective data of known quality to support a decision regarding the regulatory status and management requirements of the thorium nitrate stockpile. The sampling task was performed by RWE NUKEM, with oversight by ORNL, and the data quality objective (DQO) process was used to establish the quality and quantity of data required to satisfy decision-making needs. The data generated are compared and validated by comparison with the limits required by RCRA and by the NTS WAC.

Each lot from the stockpile was sampled (see Table II). The thorium nitrate is not a heterogeneous material; it was manufactured to be within a precise range of purity. Within each lot, the process used to make the thorium nitrate was exactly the same; therefore, the material within each drum of each lot is representative of that lot. Overall sampling accuracy refers to the closeness of sample results to the true value for the population being sampled. Because each lot is sampled, the accuracy of the material composition determination is increased. Analytical accuracy is measured in the laboratory by spiking samples with known concentrations of surrogates and comparing them with measured results. This is one of the quality assurance/quality control (QA/QC) parameters checked during the review of analytical data.

Table II. Storage Drums to Open, Sample, and Analyze by Country of Origin

	U.S.	France	India
Total number of drums	18,924	1,901	760
Lots at Curtis Bay	63	14	14
Lots at Hammond	50	0	0
Drums to open at Curtis Bay	63	14	14
Drums to open at Hammond	15	0	0
Drums to sample at Curtis Bay	63	14	14
Drums to sample at Hammond	8	0	0
Drums to analyze at Curtis Bay	23	13	0
Drums to analyze at Hammond	8	0	0

Samples were collected from the indicated number of lots and sent to the laboratory for analysis or archived for future use. At Hammond, samples from 8 lots were sent to the laboratory. At Curtis Bay, 23 samples from drums filled with thorium nitrate of domestic origin were sent for analyses; and samples from 13 drums filled with thorium nitrate of French origin and 10 drums filled with thorium nitrate of Indian origin were sent for analyses.

The photographs in Figure 3 show some of the facilities and activities associated with sampling the thorium nitrate stockpile.

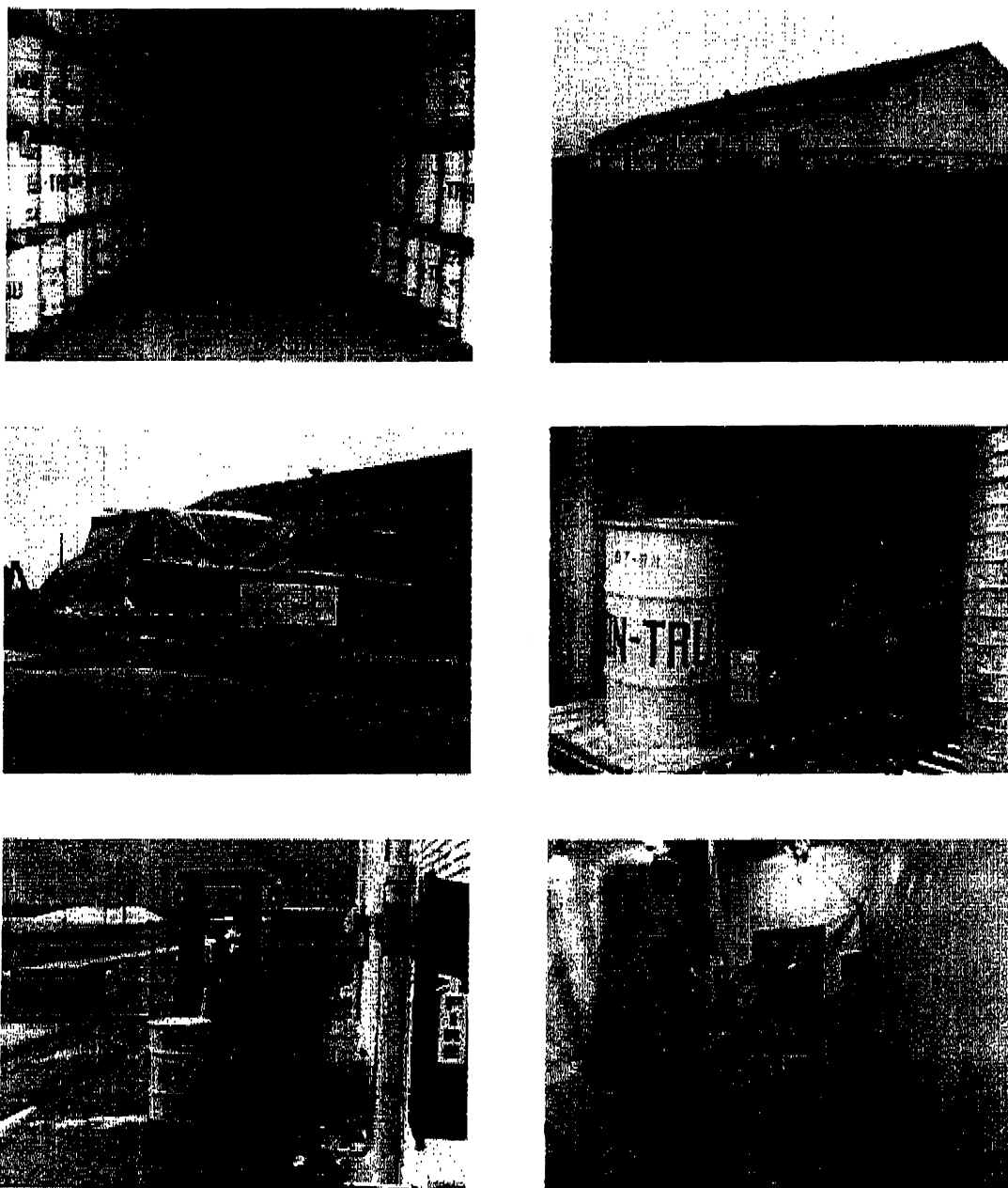


Fig. 3. Thorium nitrate stockpile facilities and drum sampling activities.

To ensure sample identification, a label containing the following information was affixed to each bottle at the time of collection: site name (Curtis Bay or Hammond); drum identification, including origin and lot number; unique sample number; date and time of collection; bottle number in the drum; and the page number of the notebook where the entries for the sample were made.

The analyses that were performed on the thorium nitrate material and the required analytical methods are compiled in Table III. Because of the expected matrix interferences, large dilutions were required. However, the laboratory reported the RCRA elements (Ag, As, Ba, Cd, Cr, Hg, Pb, and Se) with a detection limit after dilution, allowing the comparison with the RCRA limits for these elements. The detection limits after dilution did not exceed 50 mg/kg for Ag, As, Cr, or Pb; 10 mg/kg for Se and Cd; and 1000 mg/kg for Ba.

Table III. Requirements for Thorium Nitrate Samples

Analytical parameter	Analytical methods	Sample container ^a	Sample quantity ^b	Preservation	Holding time
Metals/ inorganics ^c	SW846-6020, -7471	500-mL wide-mouth HDPE bottle	Fill sample jar completely or one core sample	None	Mercury 28 days; other metals 180 days
Oxidizer test	United Nations publication ST/SG/AC.10/11.Rev.3	500-mL wide-mouth HDPE bottle	Fill sample jar completely or eight core samples	None	Indefinitely
Radionuclides (thorium, uranium)	Gamma spectroscopy (method consistent with EPA 901.1 method)	500-mL wide-mouth HDPE bottle	Fill sample jar completely or one core sample	None	Indefinitely
Archived	Any test deemed necessary	500-mL wide-mouth HDPE bottle	Fill sample jar completely or two core samples	None	Indefinitely

^aHDPE: high-density polyethylene.

^bA core sample is defined as having a height of 5 cm (2 in.) and diameter of 3.8 cm (1.5 in.).

^cMetals first interest were Ag, As, Ba, Be, Cd, Cr, Ni, Pb, Sb, Se, Th, Tl, U, V, and Zn. Other elements of interest were Al, B, Ca, Co, Cu, Fe, Mg, Mn, Mo, Na, Si, Sr, and Zr.

To confirm the detection limit achievable in the thorium matrix, an analysis was performed on a surrogate containing a standard of thorium at the same concentration as that in the sample, spiked with all the other elements at the concentration at which they are reported. This control is independent of all the regular QC associated with U.S. Environmental Protection Agency (EPA) Method SW846-6020.

The analyses followed EPA Method SW846-6020. The moisture content as well as the pH of the sample was measured and reported. All the QA/QC requested in SW846 was met. Also, the samples were analyzed for mercury according to EPA Method SW846-7471 by using cold vapor atomic absorption analysis. As for the other metal analyses, all the QA/QC included in the method was met; and any deviations were reported and documented.

The oxidizer test is critical for the project. This test was conducted according to all directives provided in the "Classification Procedures, Test Methods and Criteria Relating to Oxidizing Substances of Division 5.1" which was issued by the United Nations in 1999 as part of the *Recommendations on the Transport of*

Dangerous Goods—Manual of Tests and Criteria (Publication ST/SG/AC.10/11/Rev.3). The results of the oxidizer test varied depending upon the moisture content of the sample and the relative humidity in the room in which the experiment took place. Therefore, the sample was prepared just before testing.

The sample procedure in ST/SG/AC.10/11/Rev.3 raises the issue of friability, but it does not provide a quantitative definition of friable. This issue was investigated; and, in consultations with United Nations transportation officials and U.S. Department of Transportation staff, it was determined that there is no formal regulatory definition of "friable" as this word is used in paragraph 34.4.1.2.6 of the *United Nations Manual of Tests and Criteria*. However, it was agreed that a reasonable definition is as follows: During typical transport, a material is

- friable if more than 10% of the particles created are less than 500 μm in diameter,
- not friable if less than 5% of the particles created are less than 500 μm in diameter, and
- likely not friable if less than 10% of the particles created are less than 500 μm in diameter, but setting the limit closer to 5% provides a greater margin of safety.

After manually checking the friability of the Hammond samples received at the Southwest Research Institute (SWRI), ORNL decided to treat the material as non-friable and justified that decision by weighing both the material received intact and the material present as powder after transportation to the laboratory. It was a conservative approach which considered the total weight of all the loose material as the percentage of powder instead of the fraction that was less than 0.5 mm in diameter as called for in the UN method. Even then, none of the domestic samples consisted of a fraction larger than 10% powder. Therefore, none of the samples were friable according to the UN criterion. This conclusion is supported by the analytical results data documented in the project results report.

The samples were tested by SWRI consistent with ST/DG/AC.10/11/Rev.3 to assess if they present the characteristic of being an oxidizer. None of the 53 samples tested had a mean burn time less than that of the 3:7 reference mixture; therefore, no sample is an oxidizer according to the DOT definition.

A total analysis of the thorium nitrate material was performed by using a combination of inductively coupled plasma-atomic emission spectroscopy (ICP-AES) and mass spectroscopy (ICP-MS). The data provided information on the purity of the material as well as the possibility of being characteristically toxic per EPA definition. The thorium nitrate was found to be pure with a maximum impurities being less than 2500 mg/kg or 0.25%.

The concentration of thorium was found to be between 42 and 51.8 wt % for the domestic origin, 43.4 and 52.9 wt % for the French origin, and 45 to 53.3 wt % for the Indian origin. Sodium was found to be the largest impurity in the domestic material stored at Hammond Depot with a maximum concentration of ~2500 mg/kg. The domestic material from Curtis Bay Depot did not show such high levels of sodium with the maximum found at 156 mg/kg in only one sample. Barium was the second largest impurity found in three of the domestic material with a maximum of 647 mg/kg. Lead, thallium, manganese, sodium, and silicon were also measured in some samples at very low concentration.

The French thorium nitrate contained sodium and uranium in all the samples analyzed at maximum concentrations of 819 and 46 mg/kg respectively. Chromium, lead, and nickel were found in some samples at concentration lower than 16 mg/kg.

The Indian material contained aluminum at the maximum concentration of 467 mg/kg in nine of the 10 samples analyzed. Other impurities detected were barium (max. 19.6 mg/kg), copper (max. 14 mg/kg), lead (max. 5 mg/kg), silicon (max. 120 mg/kg), and sodium (max. 119 mg/kg).

From these results, it was proved that the thorium nitrate material from all origins was not RCRA characteristic for toxicity.

The material was also analyzed for radiological content using gamma spectrometry on all samples and alpha spectrometry on about one-third of them. The results obtained by alpha spectrometry are confirming the ICP analyses for the thorium content of the samples. However, because uranium is undetected or near the detection limit, the comparison between the two techniques is not as good for the uranium content. The gamma spectra were complex to interpret because of the large number of interference caused by radioisotopes such as ^{228}Ac that have a very large number of peaks.

MODELING FOR DISPOSAL

NTS was used as a location to model for the disposal of the thorium nitrate stockpile in an as-stored configuration. It was assumed that the thorium nitrate would remain in the present drums, and no chemical processing would be required.

The characteristics of the thorium nitrate stockpile were evaluated against the NTS WAC. Two characteristics were identified that may not be in conformance with the NTS WAC: hazardous character and activity concentration. The results from the sampling and analysis project described above established that the thorium nitrate does not contain RCRA metals at levels sufficient to be categorized as having D004-D011 character. Additionally the thorium nitrate does not meet the criteria for a DOT oxidizer; hence, it does not have D001 character.

The thorium activity concentration exceeded NTS action levels by more than a factor of three. After discussions with NTS personnel, it was determined that the content of ^{226}Ra and its progeny ^{222}Rn is considerably more important than the ^{232}Th activity concentration. Emissions of radon, in particular ^{222}Rn , from the ground surface above the burial cells are carefully monitored at NTS and must be less than NTS performance criterion, $0.74 \text{ Bq/m}^2\text{-sec}$ ($20 \text{ pCi/m}^2\text{-sec}$), at 1000 years after the disposal cell is closed.

Using the NTS deterministic performance assessment model for radon emissions, the emissions were computed at 100, 1000, 10,000, and 100,000 years after disposal. It was found that the radon emissions, specifically ^{222}Rn emissions, from the thorium nitrate stockpile would reach a maximum shortly before 10,000 years. The half-life of the ^{220}Rn progeny of ^{232}Th is much too small for ^{220}Rn to contribute measurably to the radon emissions from the surface of the ground [$t_{1/2}(^{220}\text{Rn}) = 55.6 \text{ s} = 6.44 \times 10^{-4} \text{ d}$ versus $t_{1/2}(^{222}\text{Rn}) = 3.8235 \text{ d}$]. It was also determined that the thorium stockpile must be buried more deeply than typical uranium waste to conform to the NTS performance criterion for radon emissions.

When the emissions were first modeled, there were no recent measurements of the ^{230}Th content of the stockpile. Therefore, it was assumed that the ^{238}U content of the thorium ore would determine the ^{230}Th content of the stockpile. This assumption is based upon the secular equilibrium of ^{238}U in the ore, and the partitioning of its progeny ^{230}Th with the ^{232}Th .

The ^{238}U levels in the ore were taken from published average values for the domestic, French, and Indian portions of the thorium nitrate stockpile. Based upon historical evidence collected by ORNL, each of the stockpile portions was produced from a separate ore body.

The results from emissions modeling using the ^{238}U concentrations in the ore were that the radon (^{222}Rn) emissions at 1000 years after disposal would be less than $0.01 \text{ Bq/m}^2\text{-sec}$ ($0.3 \text{ pCi/m}^2\text{-sec}$); at 10,000 years after disposal, the emissions would be about $0.03 \text{ Bq/m}^2\text{-sec}$ ($0.7 \text{ pCi/m}^2\text{-sec}$). From the modeled emissions, it was concluded that the radon emissions from the buried thorium nitrate stockpile would be less than NTS performance criterion.

After the sampling and analysis results became available, the ^{222}Rn emissions were recomputed. The measured ^{230}Th concentrations were higher than the values obtained from published ^{238}U concentrations. The newer modeled radon emissions are less than $1.2 \text{ pCi/m}^2\text{-sec}$ at 1000 years after disposal and less than $3.0 \text{ pCi/m}^2\text{-sec}$ at 10,000 years after disposal. Even with the recently measured ^{230}Th concentrations, the modeled radon (^{222}Rn) emissions are substantially below the NTS performance criterion, $20 \text{ pCi/m}^2\text{-sec}$.

CONCLUSIONS

Life-cycle analyses of a range of long-term management options were conducted. Disposal of the thorium nitrate inventory as a containerized low-level waste (without pre-treatment and/or processing) is the best option based on relative comparisons against the criteria. The characterization work scope completed in 2002 included sampling over 100 drums for RCRA (chemical) and radiological analyses. The thorium nitrate stockpile material does not exhibit hazardous characteristics, including classification as a DOT oxidizer (equivalent to RCRA D001 ignitability characteristic) and/or metals content (sufficient for RCRA D004-D011 toxicity characteristic). Sampling and analytical results support the conclusion that DNSC's thorium nitrate stockpile qualifies to be classified as low-level waste. The NTS was utilized as a case study for the disposal site. Special radiological analysis, waste acceptance, and certification requirements were evaluated for potential disposal program implementation.

FOOTNOTE

*A number of reports were prepared as a result of Phase 1 activities. See, for example, *Thorium Nitrate Material Inventory Definition Report* (ORNL/TM-2000/163, Oak Ridge National Laboratory, Oak Ridge, Tenn.), which identifies and documents the thorium nitrate material origination and processing history; documents thorium nitrate characteristics the demand for and application of thorium nitrate in the thorium fuel cycle. Also, in the *Executive Summary Report for the Thorium Nitrate Stockpile Stewardship and Disposition Project* (ORNL/TM-2001/14), generalized alternatives are defined and analyzed: (1) disposal of thorium nitrate; (2) store long-term as thorium nitrate; (3) process to convert the thorium nitrate to a form more suitable for disposal; and (4) process to convert the thorium nitrate to a form more suitable for long-term storage. Technical information and data describing viable long-term management and disposition options for the thorium nitrate stockpile are documented. Processing technologies, regulatory requirements, risks, potential environmental impacts, funding, and schedule requirements for specific options are defined.