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Title: Plutonium Packaging

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Intended for: ADPSM Plutonium Engineering Lecture, June 14, 2017

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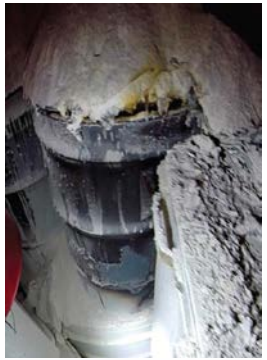
Plutonium Packaging

ADPSM Plutonium Engineering Lecture Series



Paul H. Smith

June 14, 2017

Los Alamos NATIONAL LABORATORY EST. 1943

Plutonium Handbook (2nd ed.) Chapter 32, Plutonium Packaging and Storage, Veirs, et. al.

NNSA National Nuclear Security Administration
U.S. Department of Energy's NNSA

Plutonium Packaging Overview

- Mission
- Properties
- Failures
- Regulations
- Challenges
- Acknowledgements

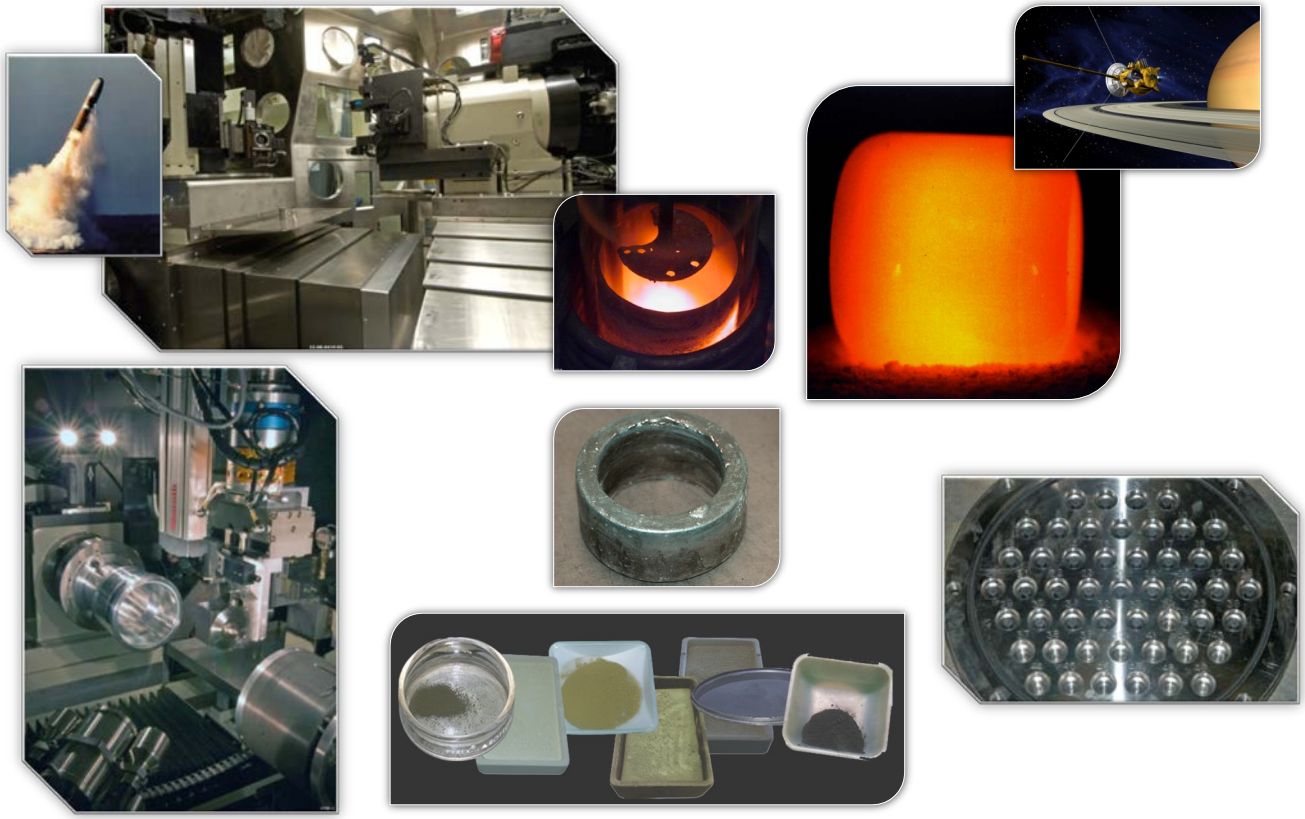
Plutonium Mission

Plutonium Science and Manufacturing

Mission

We deliver products

- Build New Pits/ Detonators
- Surveillance Report
- Disassemble Pits
- Fissile Material Disposition
- Purified Pu Oxide and Metal
- Fuels Development
- Heat Sources



Plutonium is central to all of these missions

The “Primary” Plutonium Mission

PF-4 is primarily about the capability to manufacture plutonium pits

Cassini Solstice Mission

Mission

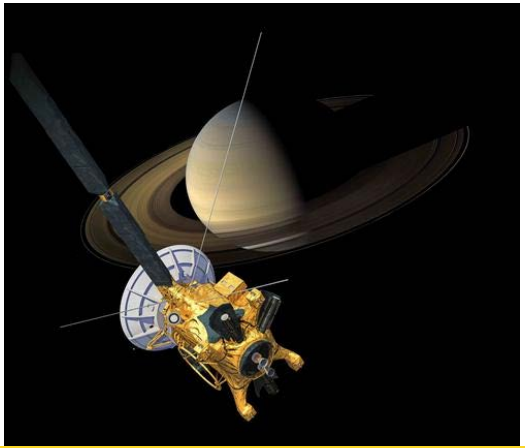
News » News Releases » 2012 » March » Oxygen Detected In Atmosphere Of Saturn's Moon Dione

Oxygen detected in atmosphere of Saturn's Moon Dione

March 3, 2012

Discovery could mean ingredients for life are abundant on icy space bodies

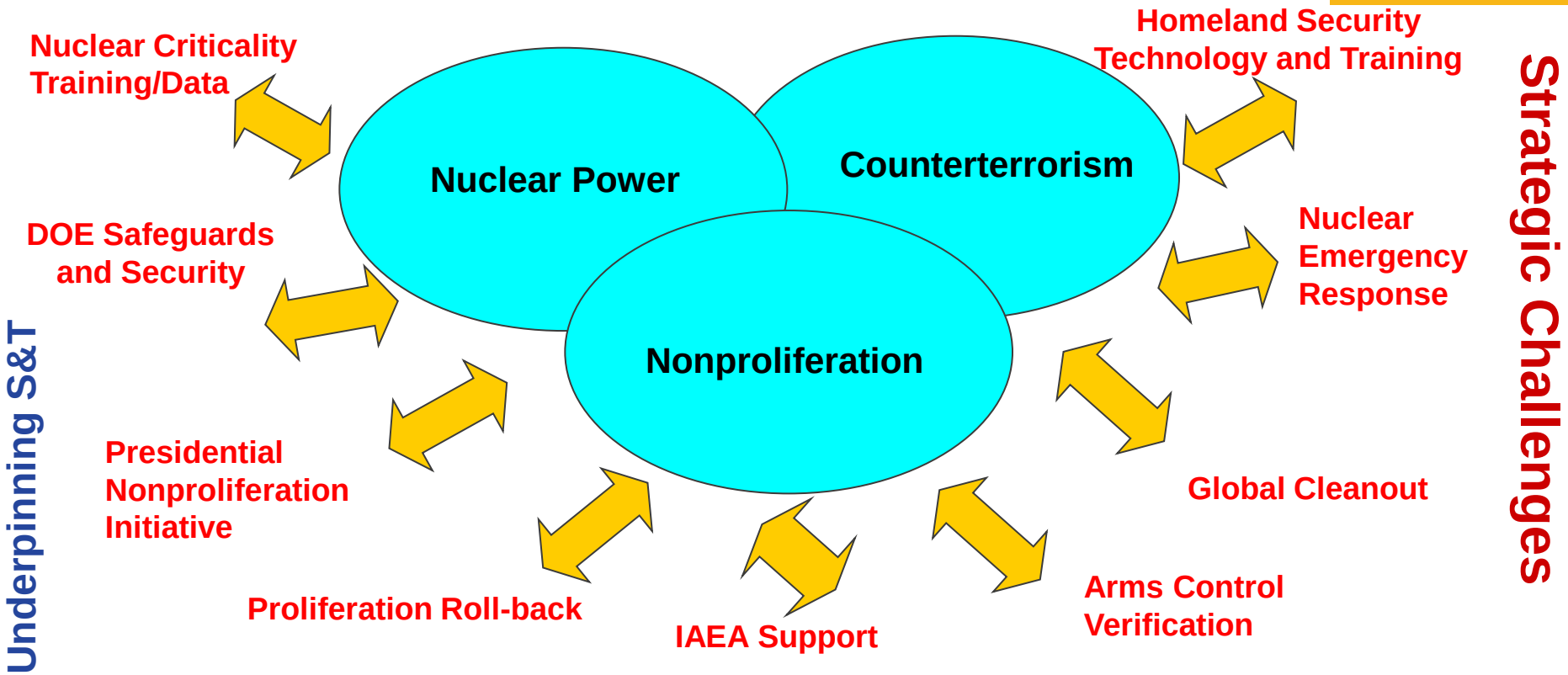
LOS ALAMOS, New Mexico, March 2, 2012—Los Alamos National Laboratory scientists and an international research team have announced discovery of molecular oxygen ions (O2+) in the upper-most atmosphere of Dione, one of the 62 known moons orbiting the ringed planet. The research appeared recently in Geophysical Research Letters and was made possible via instruments aboard NASA's Cassini spacecraft, which was launched in 1997.



Space applications depend on the heat generated by Pu-238

Nuclear Engineering and Nonproliferation Division: Nonproliferation - Key to Global Security

Mission



Chemistry, nuclear physics, solid state physics, high energy physics, materials science, information science, statistics, mathematics, critical experiments, theoretical physics, earth science, space science, nanotechnology

Plug: Nuclear Safeguards Summer Seminar Series

Mission

NUCLEAR SAFEGUARDS
LOS ALAMOS NATIONAL LABORATORY

Nuclear Lunches

A summer seminar and discussion series for safeguards, nonproliferation, and chemistry. All sessions are from **11:30 am-12:30 pm.**

Preliminary Schedule

Date	Room	Topic and Speakers
5/31	35-2-B125	Safeguards Overview, Rebecca Stevens (NEN-1)
6/7	35-2-B125	IAEA Inspection Measurements, Bill Geist (NEN-1), Rob Steiner (C-NR)
6/14	35-127-A101	Safeguards R&D: NuGoo and Hyperspectral Imaging, Vlad Henzl (NEN-1)
6/21	35-2-B125	Safeguards R&D: LIBS/LA-ICP-MS, Beth Judge, Keri Campbell (C-CDE)
6/28	35-2-B125	Fuel Cycle Safeguards: Uranium Mining, Milling, and Conversion. Heather Hawkins-Erpenbeck (NEN-1), Lisa Colletti (C-AAC)
7/5	35-2-B125	Fuel Cycle Safeguards: Enrichment, Scott Demuth (NEN-5)
7/12	35-2-B125	Fuel Cycle Safeguards: Spent Fuel, Kate McIntosh (C-CDE), Alexis Trahan (NEN-1)

Opportunity to get a glimpse into the nuclear safeguards world

Scope of Plutonium Packaging and Storage

Mission

- Plutonium in-process at nuclear weapons laboratories
  - Several metric tons
  - High-purity plutonium dioxide, metals and alloys, compounds, and processing residues
  - Wide range of radiation activity, from weapons-grade material to pure ^{241}Am or 90% ^{238}Pu
  - DOE Manual 441.1-1 Nuclear Material Storage
  - Packaging is in containers that are easily opened and closed
  - Storage duration: 1-40 years

- Plutonium-bearing materials packaged for storage for up to 50 years with minimal surveillance
  - Tens of metric tons in the United States and Russia
  - Metals and oxides containing at least 30 wt % plutonium plus uranium
  - DOE's 3013 Standard: Stabilization, Packaging, and Storage of Plutonium-Bearing Materials
  - Storage duration: many years or decades

- Plutonium from reprocessed spent fuel from commercial reactors
  - Over 100 metric tons
  - High-purity plutonium dioxide
  - High radiation activity, increases with age as ^{241}Am grows in, high temperature
  - Packaging requirements vary
  - Packaging is two nested, welded stainless steel containers
  - Storage duration: many years or decades

Safe plutonium storage is essential to national security and energy missions

Magnitude of 3013 Storage

Mission

A single container failure is unacceptable

Source of 3013 Containers	
	# 3013s
Hanford	2257
LANL	50
LLNL	198
RFETS	1888
SRS	902
	5295

Current Storage Configuration…3013s stored at Savannah River Site in 9975 Type B shipping containers

DOE's 3013 Standard requires a surveillance program which includes both non destructive and destructive examinations (NDE and DE)

Plutonium Properties

Criticality Safety for ^{239}Pu

Properties

- Critical mass at full density for ^{239}Pu
  - Plutonium metal 5.0 kg
  - Plutonium dioxide10.2 kg
- All packaging and storage locations require criticality safety evaluations
  - Mass, chemical form, shielding, hydrogen content, neutron poisons, etc.
  - Spacing, storage arrays
  - Normal and off-normal conditions
- Potential for water ingress
  - Water is a neutron moderator, increases probability of causing fission
  - Water solubility of plutonium form comes into play

Criticality safety must be included in the container design process

Plutonium Radiation Considerations

Properties

- α, β, γ, and neutron radiation will be present depending on isotopic mixture
- Inhaled airborne particles are used to define threshold amounts for packaging
- Containers must be designed to limit the committed effective dose equivalent (CEDE) of a worker to less than 50 mSv (5 rem) in:
  - Normal handling conditions
  - Off-normal handling conditions, e.g., dropped container
- Continuous Air Monitor (CAM) alarm
  - Airborne contamination is present
  - Exit room immediately

Plutonium containers must mitigate the risk of plutonium inhalation

Radioactive Properties of Plutonium Isotopes and Other Common Actinides

Properties

Nuclide	Half-life,[a] yr	AMU, g/mol	SA,[b] TBq/g	E,[c] MeV/nt	SP,[d] W/kg	A_2,[b] TBq	A_2 (g)
^{234}U	2.455×10^5	234.040946	2.3×10^{-4}	4.8587	1.793×10^{-1}	6.0×10^{-3}	
^{235}U	7.038×10^8	235.043923	8.0×10^{-8}	4.6891	6.011×10^{-5}	Unlimited	Unlimited
^{236}U	2.342×10^7	236.045562	2.4×10^{-6}	4.5723	1.754×10^{-3}	6.0×10^{-3}	
^{238}U	4.468×10^9	238.050783	1.2×10^{-8}	4.2691	8.512×10^{-6}	Unlimited	
^{237}Np	2.144×10^6	237.048166	2.6×10^{-5}	4.9529	2.067×10^{-2}	2.0×10^{-3}	
^{238}Pu	8.77×10^1	238.049553	6.3×10^{-1}	5.593	5.68×10^2	1.0×10^{-3}	1.6×10^{-3}
^{239}Pu	2.411×10^4	239.052156	2.3×10^{-3}	5.2442	1.930	1.0×10^{-3}	4.4×10^{-1}
^{240}Pu	6.561×10^3	240.053807	8.4×10^{-3}	5.2559	7.077	1.0×10^{-3}	
^{241}Pu	1.4329×10^1	241.056844	3.8	0.0054	3.3	6.0×10^{-2}	
^{242}Pu	3.75×10^5	242.058736	1.5×10^{-4}	4.9855	1.176×10^{-1}	1.0×10^{-3}	
^{241}Am	4.327×10^2	241.056822	1.3×10^{-1}	5.6379	1.146×10^2	1.0×10^{-3}	7.9×10^{-3}

Plutonium Handbook (2nd ed.) Chapter 32, Plutonium Packaging and Storage, Veirs, et. al.

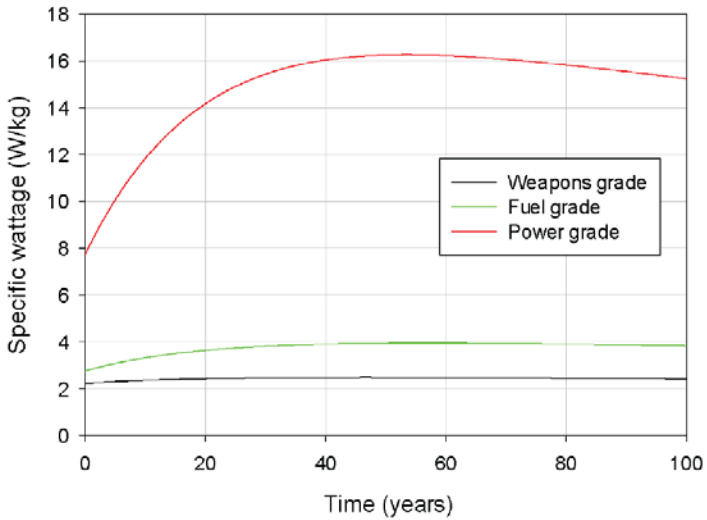
Grades of Plutonium

Properties

Isotopic Mixtures for Pu Grades

Nuclide	Weapons grade	Fuel grade	Power grade
^{238}Pu	0.05%	0.1%	1.0%
^{239}Pu	93.50%	86.1%	63.0%
^{240}Pu	6.00%	12.0%	21.0%
^{241}Pu	0.40%	1.6%	12.0%
^{242}Pu	0.05%	0.2%	3.0%
^{241}Am	0.00%	0.00%	0.00%
Initial SP, W/kg	1.93	2.53	3.15
Maximum SP, W/kg	1.93	2.81	18.5

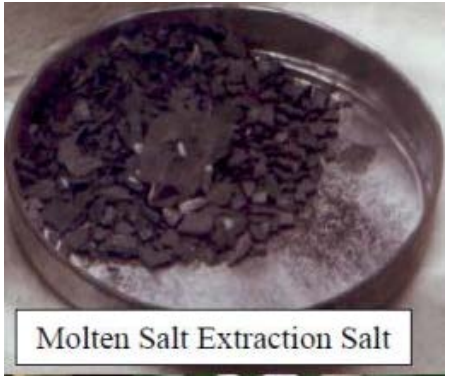
Change in Specific Wattage with Time

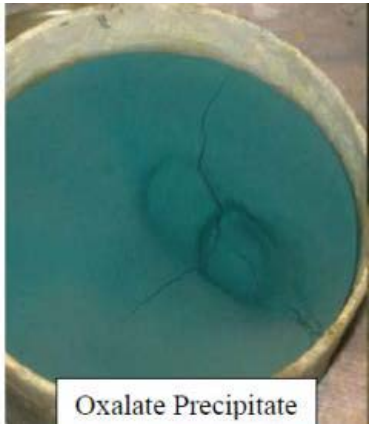


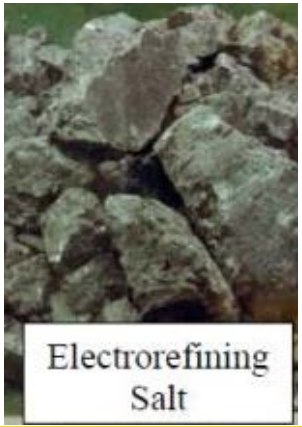
Plutonium Handbook (2nd ed.) Chapter 32, Plutonium Packaging and Storage, Veirs, et. al.

Plutonium Compounds and Residues

Properties







Chemical form of Pu must be compatible with container components

Plutonium Metal Can Spontaneously Ignite in Air

Properties

Plutonium metal is generally packaged in hermetically sealed inners

Plutonium Package Failures

Container Failure Case Studies

Failures

- Fifteen instances of container failure resulting in contamination of workers and equipment have been identified to have occurred between 1969 and 1999
- In 1993 one worker was contaminated while handling a package containing 2.5 Kg Pu metal casting
- On November 10, 2011 a failed ZPPR container with metal resulted in worker uptakes
- On August 5, 2003, in the plutonium facility at Los Alamos National Laboratory, a radiological release of Pu-238 occurred
- On December 19, 2005 contamination was released during the annual inventory of the vault in the Plutonium Facility at Los Alamos National Laboratory resulting in contamination
- On February 14, 2014 a container failure occurred at the Waste Isolation Pilot Plant (WIPP) that resulted in contamination of 16 workers and release of plutonium and americium to the environment

Plutonium Handbook (2nd ed.) Chapter 32, Plutonium Packaging and Storage, Veirs, et. al.

Plutonium Packaging Failure Examples

Failures

Pu Metal, LANL, 1993

Pu-238 Residue, LANL, 2003

Pu Oxide, LANL, 2005

ZPPR, Idaho, 2011

Pu/Am Residue, WIPP, 2014

We are still learning from plutonium package failures

Plutonium Packaging Failure Configurations

Failures

Pu metal in steel cylinder
2 layers plastic
Taped slip lid

Pu-238 corrosive rags
Taped slip lid with seam

Fuel-grade Pu-239
Taped slip lid with seam
Plastic inner container

Fuel-grade Pu-239 metal
Welded steel plate

Pu-239 residue, high Am
Nitrate salts, organic kitty litter

Plutonium Handbook (2nd ed.) Chapter 32, Plutonium Packaging and Storage, Veirs, et. al.

Evaluation of Container Failures against the Stored Material Characteristics Requirements from DOE M 441.1-1

Failures

M441.1-1 Scope of Materials Criteria	ZPPR	Pu-238	Fuel	WIPP
Explosion Sensitive and/or Flammable Materials	Y	Y	Y	N
Gas Generation	Y	N	N	Y
Incompatible materials	Y	N	N	N
Physical and Chemical Form	Y	N	N	N
Moisture Content	Y	Y	Y	N
Pyrophoricity	Y	Y	Y	Y
Radioactive Decay Heat	Y	N	Y	Y
Radiation Fields	Y	N	Y	Y
Solution	Y	Y	Y	N

If the package met the criterion it was given a Y (for yes) and if the package did not meet the criterion it was given an N (for no).

Plutonium Handbook (2nd ed.) Chapter 32, Plutonium Packaging and Storage, Veirs, et. al.

Evaluation of Container Failures against Packaging Design Criteria and Performance Objectives Requirements from DOE M 441.1-1

Failures

M441.1-1 Packaging Criteria	ZPPR	Pu-238	Fuel	WIPP
Corrosion resistant	Y	N	N	Y
Thermal degradation resistant	Y	N	N	Y
Radiation resistant	Y	N	N	Y
Oxidative expansion accommodation	N	Y	Y	Y
Pressurization	Y	N	N	N
Incompatible and Pyrophoric Materials	Y	N	N	N
Filter Performance	Y	Y	Y	N
Design Life	N	N	N	Y
Container Closure	Y	N	N	Y
Design Release Rate	Y	N	N	Y
Design Qualification Release Rate	Y	N	N	Y
Non-Destructive Examination	Y	Y	Y	Y
Quality Assurance	Y	N	N	Y
Labeling	Y	Y	Y	Y
Surveillance	Y	N	N	Y

If the package met the criterion it was given a Y (for yes) and if the package did not meet the criterion it was given an N (for no).

Plutonium Handbook (2nd ed.) Chapter 32, Plutonium Packaging and Storage, Veirs, et. al.

Regulations

Defense Nuclear Facility Safety Board (DNFSB) Recommendations Drove Standardization

Regulations

- **Recommendation 94-1**
  - Resulted in 3013 standard with container requirements for long term storage (up to 50 years)
  - Interim storage requirements specified
- **Recommendation 2000-1**
  - Resulted in the development of container requirements for excess materials and a plan for the disposition of these materials
- **Recommendation 2005-1**
  - Resulted in the Nuclear Material Packaging Manual (M441.1-1) for interim storage
  - LANL Hagan style container did not meet all requirements

DOE accepted these recommendations to improve worker safety

DOE 3013 Standard and DOE Manual M441.1-1

Regulations

NOT MEASUREMENT SENSITIVE

DOE-STD-3013-2012
MARCH 2012

DOE STANDARD

STABILIZATION, PACKAGING, AND STORAGE OF PLUTONIUM-BEARING MATERIALS

U.S. Department of Energy
Washington, D.C. 20585

AREA PACK

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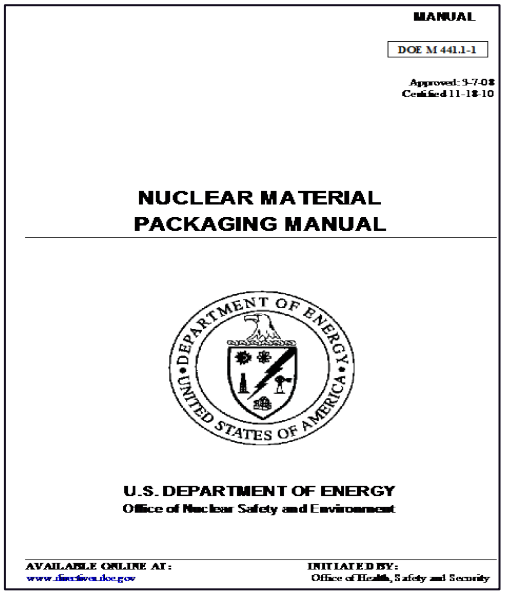


MANUAL

DOE M 441.1-1

Approved: 9-7-08
Canceled 11-18-10

NUCLEAR MATERIAL PACKAGING MANUAL

U.S. DEPARTMENT OF ENERGY
Office of Nuclear Safety and Environment

AVAILABLE ONLINE AT:
www.directives.doe.gov

INITIATED BY:
Office of Health, Safety and Security

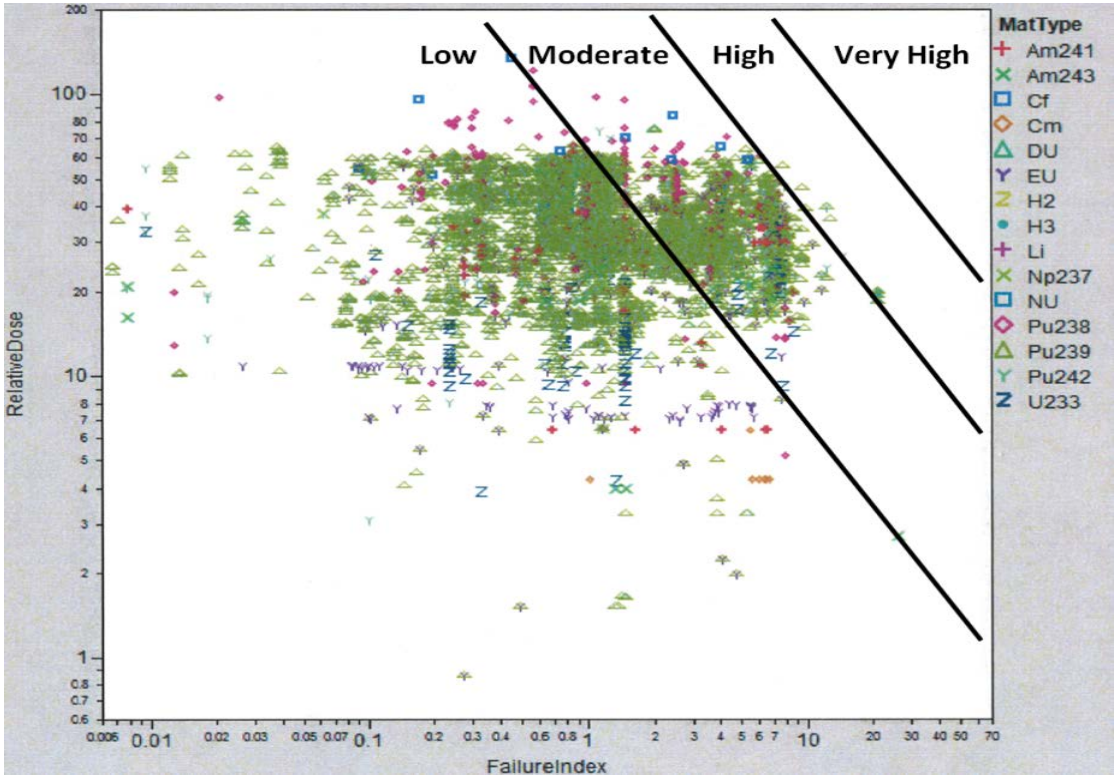
- Long Term Storage, 50 yr
- Hermetically sealed design
- Pu materials >30 wt.%

- Interim Storage, 1-50 yr
- Vented designs, easily opened/closed
- Actinides, respirable, > 0.4 g Pu

Include requirements for material characteristics and container design

Risk-Based Methodology for Repackaging

Regulations



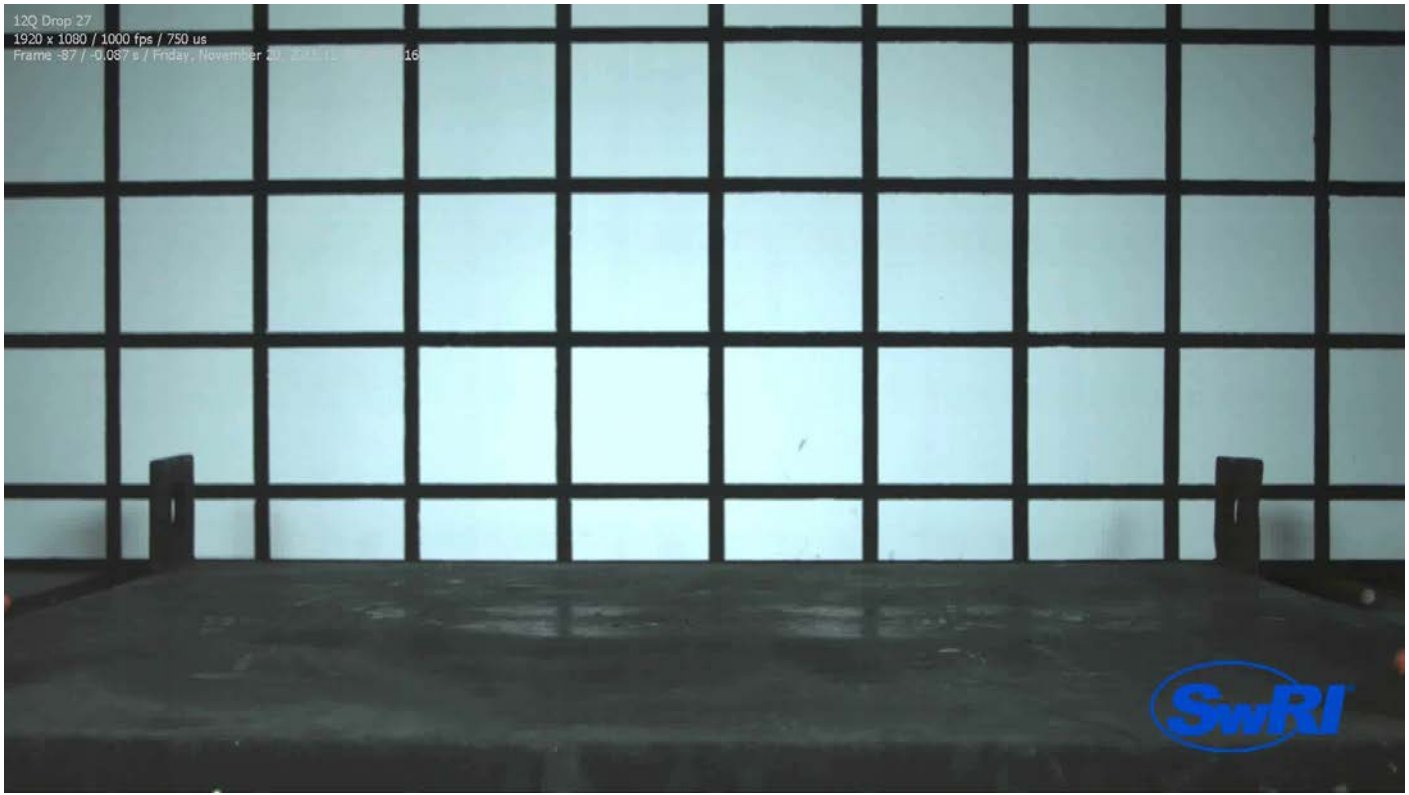
Risk = (consequence) x (probability of failure) ~ dose x age

Challenges

Test Hagan container relative to M441.1-1

Challenges

Hagan Design 1998

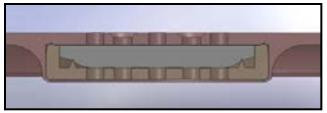


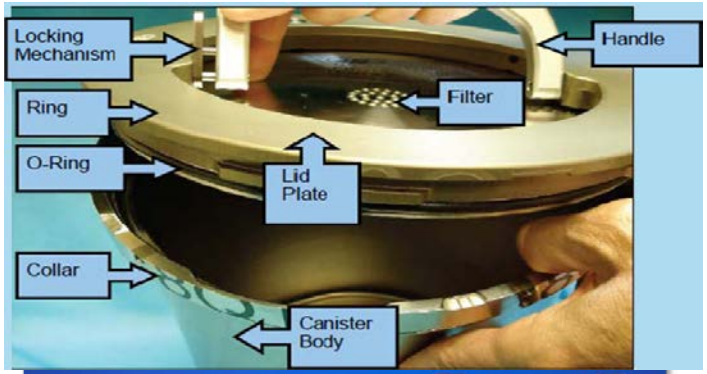
The Hagan container does not meet M441.1-1 requirements

Design Container to Meet M441.1-1

Challenges

SAVY 4000 Design and Testing





We have met this challenge with the SAVY-4000 design for 5 year design life

Demonstrate M441.1-1 Compliance at LANL

Challenges

Procured He Leak Test System (10/10) and Fully Installed in PF-4 (2/12)

Developed FilterTest System and Fully Installed in PF-4 (12/13)

Acceptance Test >1700 SAVY-4000 containers

CW Production Laser Welder Installed and Qualified (4/11, welded >1400 parts as of 2013)

Installed Bar-Code Readers in PF-4 (1/12)

Operating Procedures and Training (7/11 to present)

First Packaging of Plutonium (Begun, 8/2011
230 currently)

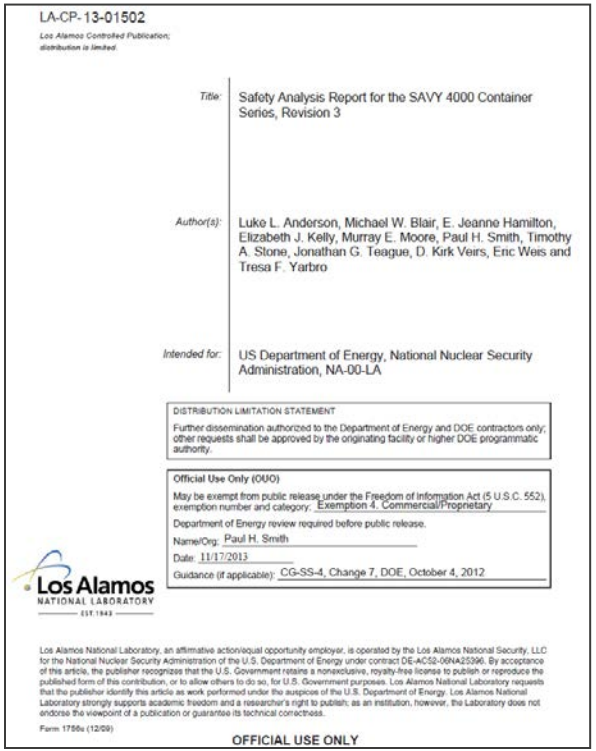
O-ring Durometer and Visualization equip. installed in PF-4 (11/2012)

Major aspect of compliance was establishing surveillance program

Demonstrate M441.1-1 Compliance at LANL (cont.)

Challenges

- Revision 0 submitted January, 2011
- Q0 Comments received May, 2011
- Revision 1 submitted March, 2012
- Q1 Comments received January, 2013
- Revision 2 submitted March, 2013
- Q2 Comments received October, 2013
- Revision 3 submitted November, 2013
- OP&T issued SER, April, 2014
- LA Field Office Approved SAR, April, 2014



LA-CP-13-01502

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Title: Safety Analysis Report for the SAVY 4000 Container Series, Revision 3

Author(s): Luke L. Anderson, Michael W. Blair, E. Jeanne Hamilton, Elizabeth J. Kelly, Murray E. Moore, Paul H. Smith, Timothy A. Stone, Jonathan G. Teague, D. Kirk Veirs, Eric Weis and Tresa F. Yarbro

Intended for: US Department of Energy, National Nuclear Security Administration, NA-00-LA

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Name/Org: Paul H. Smith

Date: 11/17/2013

Guidance (if applicable): CG-SS-4, Change 7, DOE, October 4, 2012

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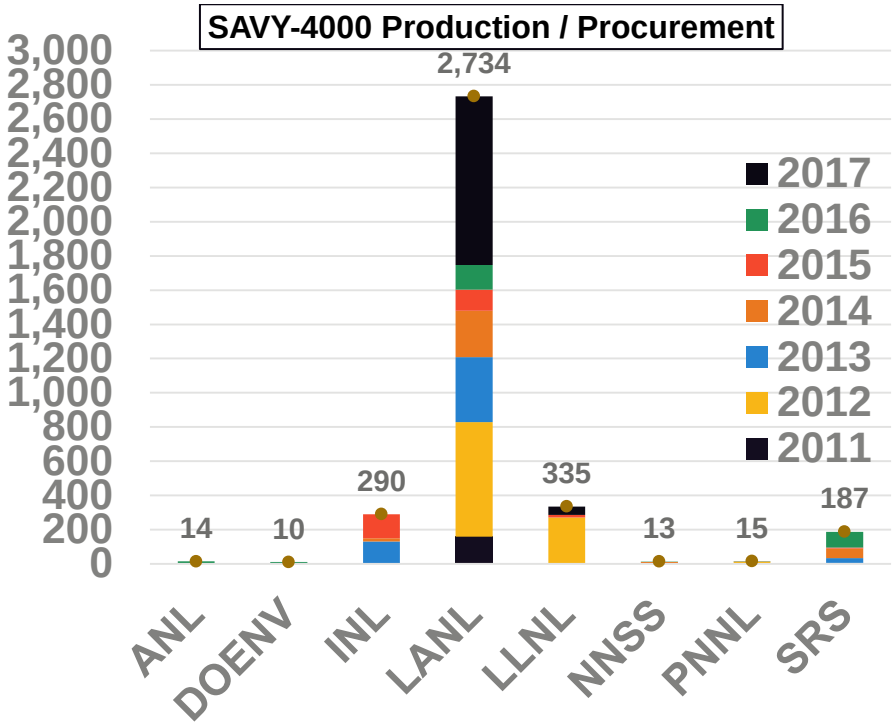
Form 1756a (12/09)

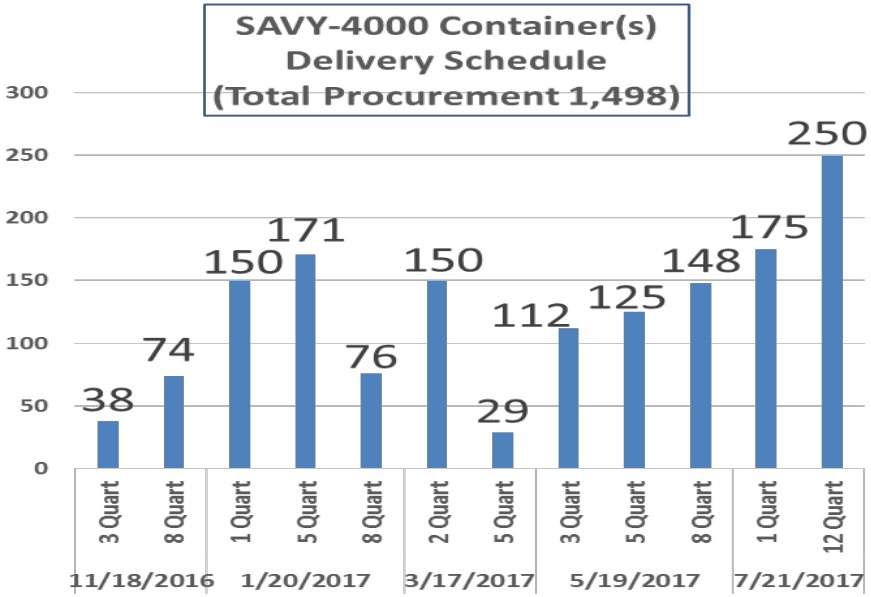
OFFICIAL USE ONLY

Safety analysis report peer reviewed by DOE Packaging and Transportation

Manufacture Containers to NQA-1 Standard

Challenges



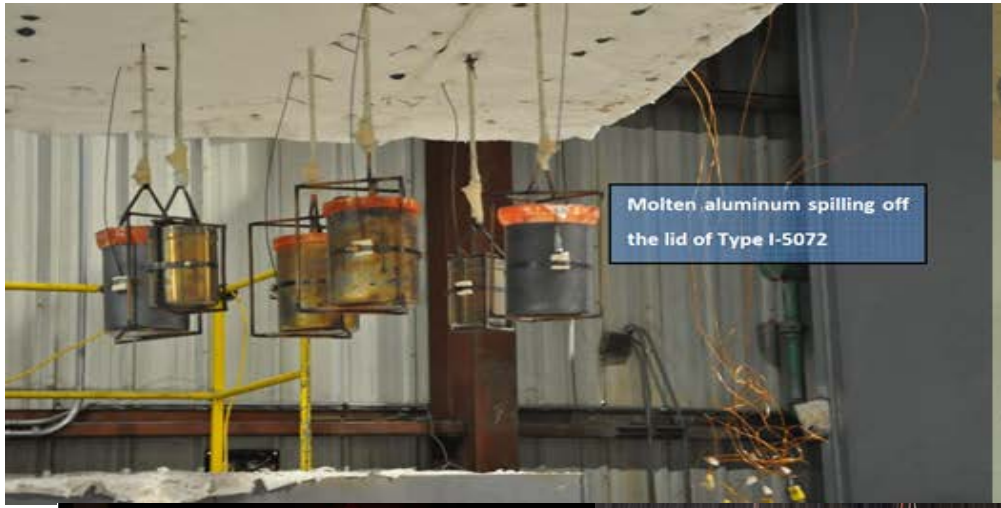


3,598 Containers Manufactured to Date **425 Remaining in Production**

Containers now being used across the DOE complex

Demonstrate MAR Reduction for SAVY in TA-55

Challenges



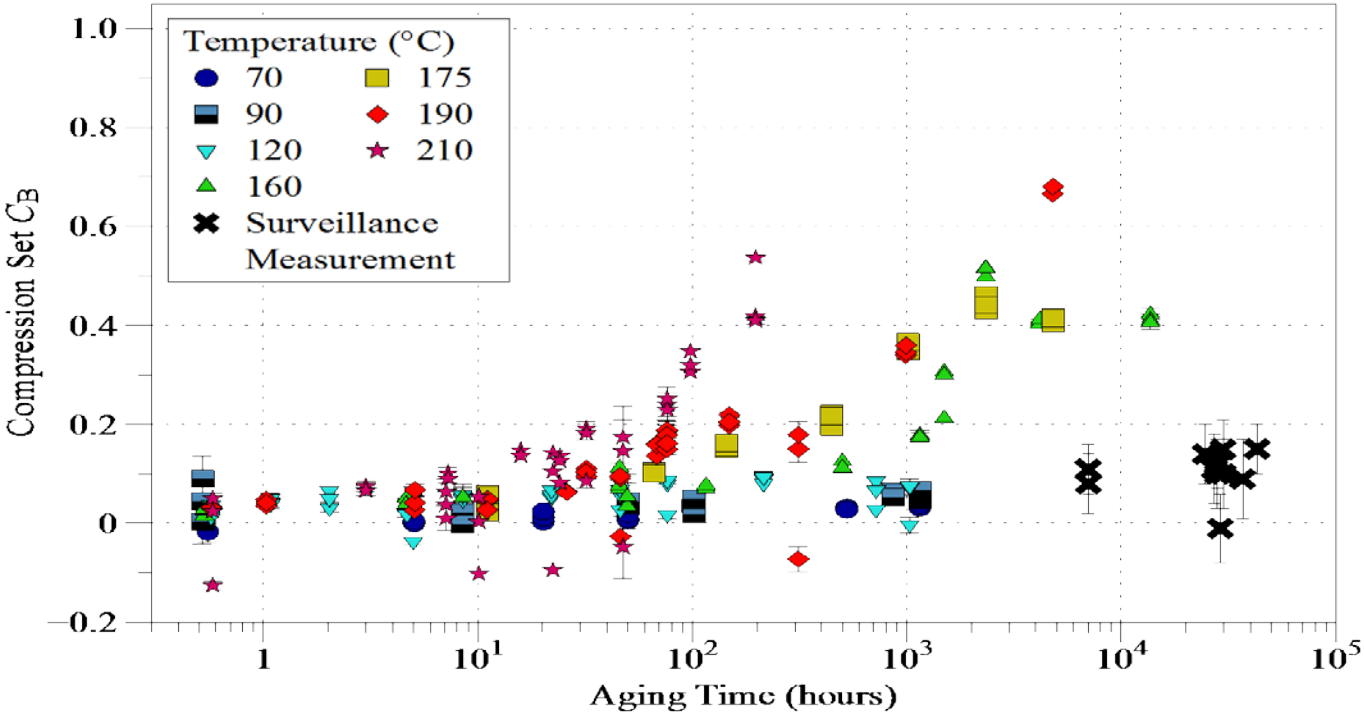
SAVY's credited to reduce MAR by a factor of 99% in PF-4 starting in 2012

Extend SAVY-4000 Design Life - Viton

Challenges

Compression set on field samples is under 20%; these data follow the same trends exhibited by the low-temperature accelerated aging samples

Time-Temperature Superposition



We can now defend a 40-year design life for the Viton o-ring

Extend SAVY-4000 Design Life – Stainless Steel

Challenges

Corrosion Observations

Hagan

SAVY-4000

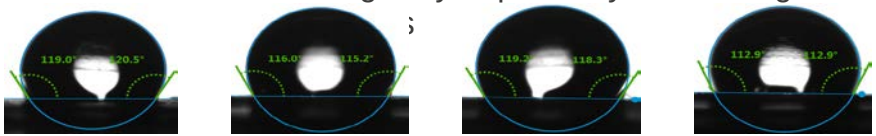
Examining severity and mechanisms: surface, pitting, stress corrosion cracking

Extend SAVY-4000 Design Life – Filter Membrane PE/PTFE Membrane Exposure to γ Radiation

Challenges

No Major Surface Changes → Maintains Hydrophobic/Waterproof Properties

- Polyethylene (PE) – Inner Side
  - Contact Angle
    - Slight Hydrophobicity Loss at Higher



Pristine | 1 Mrad | 2 Mrad | 5 Mrad

Sample Exposure Details

Modeled Exposure Time	Average Contact Angle (Degrees)
Pristine	120
1 Mrad	116
2 Mrad	118
5 Mrad	112

- Teflon (PTFE) – "Waterproof" Outer Side
  - Contact Angle
    - Mostly Maintains Hydrophobicity

Pristine | 1 Mrad | 2 Mrad | 5 Mrad

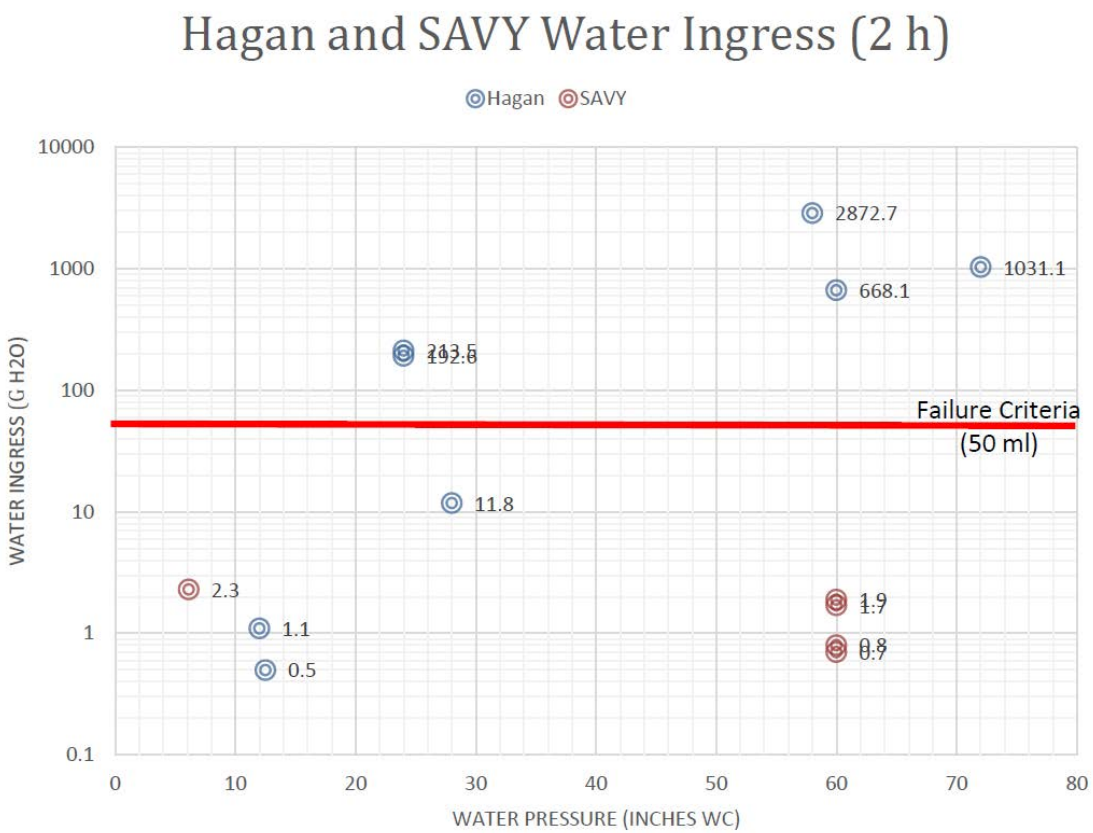
Sample Exposure Details

Modeled Exposure Time	Average Contact Angle (Degrees)
Pristine	140
1 Mrad	142
2 Mrad	146
5 Mrad	140

Membranes are expected to receive an estimated dose of 4.2 Mrad over 40 yrs

Demonstrate Water Resistance for Inside Glovebox Usage

Challenges



Hagan and SAVY containers resist water entry, <50 ml @ 12 inches WC

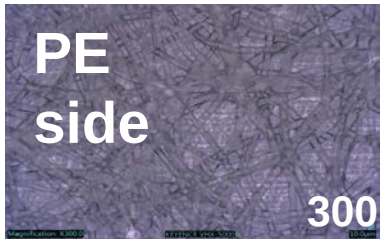
Demonstrate Ongoing Water Resistance for Inside Glovebox Usage

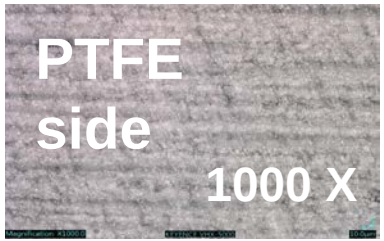
Challenges

PE/PTFE Membrane Degradation by α Radiolysis

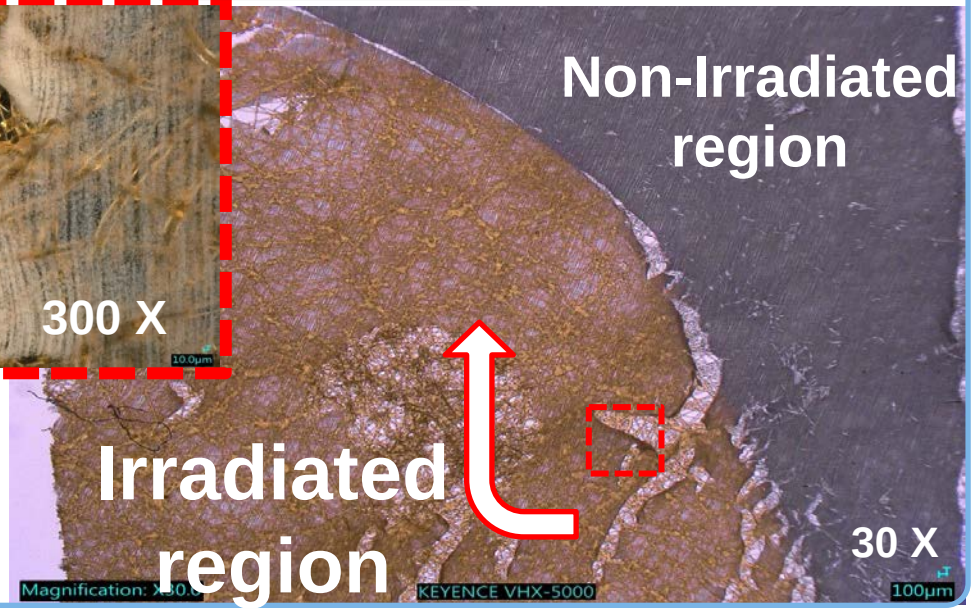
Digital Microscope Images Reveal Extensive Structural Damage

Pristine





Irradiated



Sample showed cracks and discoloration

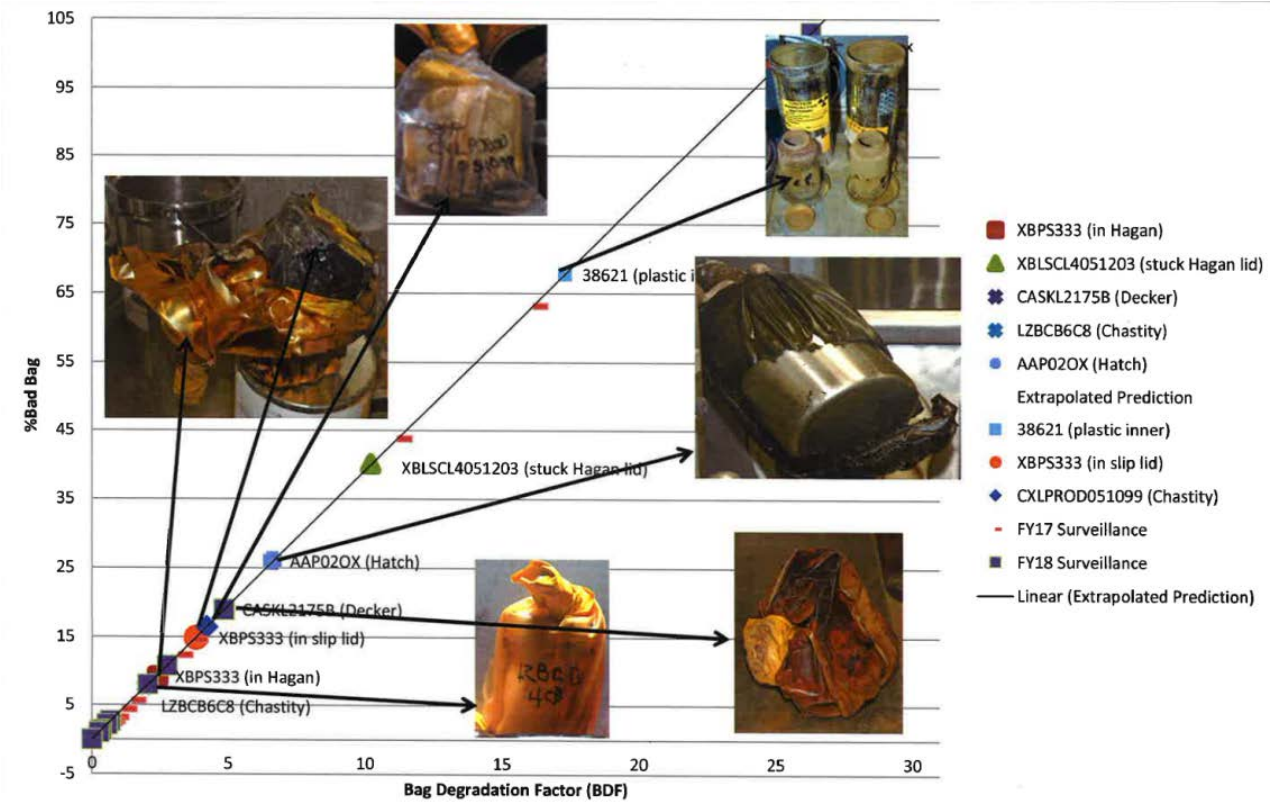
Understand the Role of PVC Bagout Bags in Corrosion

Challenges

Polyvinylchloride bagout bags are likely causing corrosion

Predict Bagout Bag Degradation Based on Survey Data

Challenges



Bag Degradation Factor = Power * Age / Radius^2

Deal with the Unexpected

Challenges

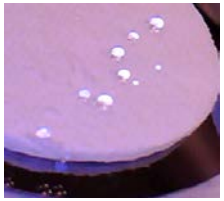
- No external contamination
- Item identified during In-Service Inspection performed as part of MR&R re-shelving operation
- White powder identified as ammonium chloride
- Ammonium chloride on the container and shelf
- Vault personnel inspected vault locations and discovered other containers exhibiting similar behavior – no external contamination
- Container was introduced into a glovebox for evaluation and testing

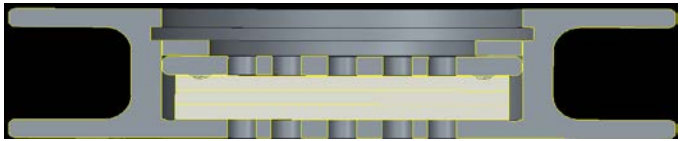
Containers continuing to function as designed

Develop Water, Radiation, Fire Resistant Filter Material

Challenges

- Funded initially by DOE Office of Environment, Health, Safety and Security, Nuclear Safety to identify a water, fire and radiation resistant filter media for nuclear material container for glove-box use
- Treated current filter media to resist water penetration
  - Meets filter efficiency, pressure drop, water resistance and hydrogen diffusion criteria
  - Remains hydrophobic after 2 hours at 450°C
- Designed and built test fixture
  - Mimics actual SAVY-4000® lid design
  - Reloadable
  - Allows variable compression on filter media





Testing PFOTS treated sample for hydrophobicity

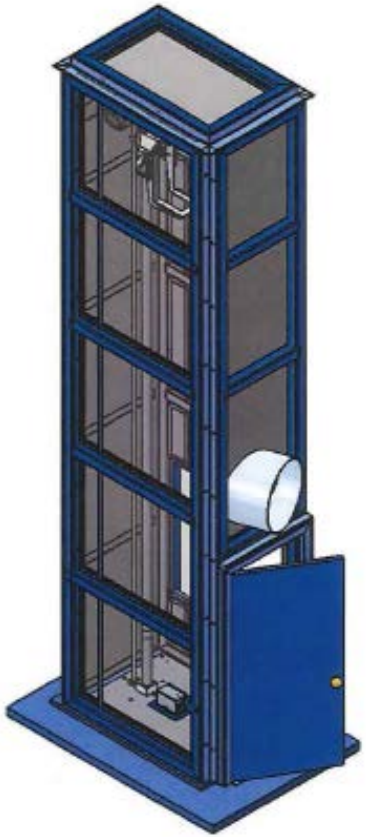
PFOTS treated fibrefrax material, max compression 1 layer of material

Filter Coating	Percent Penetration	Pressure Drop (inWC)	Water Penetration Result
1 mM PFOTS 3 coats	0.0008%	0.2	PASS
1 mM PFOTS 4 coats	0.0005%	0.336	PASS
3 mM PFOTS 3 coats	0.0004%	0.338	PASS
3 mM PFOTS 4 coats	0.0003%	0.345	PASS

New material submitted for patent, pursing licensing for technology transfer

Establish Capability to Measure Container Release Directly

Challenges



RRFMC key features:

- Remote control, up to 16 ft high
- Total container capacity 150 lb
- The drop tower will be added to existing HEPA filtered wind tunnel
- Test aerosol particles can be counted with no background interference

The Respirable Release Fraction Measurement Chamber

Design General Purpose In-Glovebox Storage Container

Challenges

- Finalizing Requirements Document (RD) before being submitted through the LANL Document Review System
  - Water Resistant before/after GB fire
  - Easy to use, cost effective
  - MAR credited
- Initial designs of a positive closure mechanism for the new container are being analyzed
  - A decision matrix tool for the closing mechanism has been developed with respect to the RD
- New In-glovebox working group established 3-21-17, meeting bi-weekly
- 10 designs currently being evaluated
- Expect to have prototypes for testing by the end of FY17

Design Criteria Decision Matrix

Design Criteria	weight 10=most critical
Water resistance	10
Fire resistance	7
Drop test effective	7
Cost wrt SAVY	8
Ease of use wrt SAYY	8
Safety/glove puncture	9
Inspectability/Surveilla	5
Weight wrt SAVY	3

Major challenge: meet multiple requirements, keep it simple, safe, cheap

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