

# Finding SNM and Homeland Security Applications

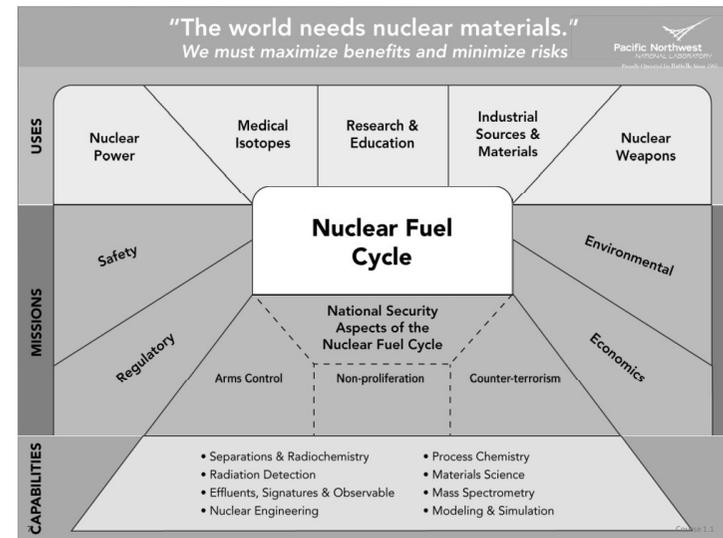
Ongoing projects



Pacific Northwest  
ANALYTICAL & CHEMICAL SERVICES  
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## The Nuclear Security Mission

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## Nuclear Threats



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<p><b>Tactical</b></p> <ul style="list-style-type: none"> <li>▪ Nuclear weapons in the hands of terrorist organizations and rogue nations</li> <li>▪ Nuclear attack on population centers, critical infrastructure, or the global supply chain</li> </ul>	<p><b>Strategic</b></p> <ul style="list-style-type: none"> <li>▪ Proliferation of nuclear technology to rogue states</li> <li>▪ Potential to upset the balance of power</li> </ul>
<p><b>International cooperation increases the ability to deter and respond to nuclear material out of regulatory control</b></p>	<p><b>Treaties and agreements lessen the risk of nuclear material and technology leaving regulatory control</b></p>

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## Security and Safeguards

- *safeguards* and *security* are generally used to describe programs **that promote the common defense and security and protect public health and safety** by guarding against theft and sabotage. **The licensee security programs and contingency plans deal with threats, thefts, and sabotage relating to special nuclear material, high-level radioactive wastes, nuclear facilities, and other radioactive materials and activities that the NRC regulates.**

## Relevant Treaties



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**Treaty on the Nonproliferation of Nuclear Weapons (NPT)**  
*technical challenge*  
 detect undeclared production and processing of nuclear material in non-weapons states prevent illicit nuclear material trafficking

**new Strategic Arms Reduction Treaty (START)**  
 new treaty entered into force on 5 Feb 2012  
*technical challenge*  
 characterize nuclear material; decipher weapons from the rest  
 integrate radiation detection with companion technologies and information barriers

**Comprehensive Nuclear Test Ban Treaty (CTBT)**  
*technical challenge*  
 monitor radioisotope emissions from (underground) nuclear tests  
 sustain and expand existing network for timely, definitive event characterization

**proposed Fissile Material Cutoff Treaty (FMCT)**  
*technical challenge*  
 not yet clear; need verification measures that don't rely on national technical means need to discover and define signatures unique to contemporary production

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<p><b>Material Production</b></p> <ul style="list-style-type: none"> <li>▪ physical protection</li> <li>▪ international safeguards</li> <li>▪ export control and regulation</li> <li>▪ undeclared production</li> </ul>	<p><b>Border and Transportation Security</b></p> <ul style="list-style-type: none"> <li>▪ screening of people and conveyances</li> </ul>
<div style="background-color: #555; color: white; padding: 10px; border-radius: 10px; display: inline-block;"> <h3>Nuclear Security Missions</h3> </div>	
<p><b>Emergency Response</b></p> <ul style="list-style-type: none"> <li>▪ searching for stolen, lost, and orphan sources</li> <li>▪ consequence management</li> </ul>	<p><b>Arms Control and Nuclear Test Monitoring</b></p> <ul style="list-style-type: none"> <li>▪ warhead counting</li> <li>▪ international monitoring system</li> </ul>

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## Sources of Interest – Threats and Non-Threats

- State-developed weapon package
- Improvised Nuclear Device (IND)
- Sub-critical portion of Special Nuclear Material (SNM)
- Radiological Dispersal Device (RDD or “dirty bomb”)
  - Conventional explosive used to drive radioactivity dispersal
- Radiation sources suitable for constructing an RDD
  
- Benign (eg, medical, industrial, or naturally occurring)
  - Can mask threat
  - Can be mistaken for threat

## Grand R&D Challenges

Detection of special nuclear material (SNM) even when heavily shielded (Shielding)



Enhanced wide area search in a variety of scenarios, to include urban and highly cluttered environments (Search)



Cost effective equipment with sufficient performance to ensure wide spread deployment (Cost)



Monitoring along challenging GNDA pathways, to include general aviation, small vessels, and in between ports of entry (Pathways)



Forensic determination of origin and/or route of interdicted materials (Forensics)



## R&D Program



Reduce the threat to national security posed by the proliferation of nuclear weapons or materials by developing the U.S. capabilities to monitor nuclear treaties, weapons development activities, and detonations worldwide.

### Nonproliferation



- SNM production capabilities
- Weapons development detection
- Operationally focused technical nuclear forensics

### Verification and Monitoring



- SNM Movement Detection
- Warhead monitoring
- International Safeguards
- Warhead Chain-of-Custody



- Produce and improve U.S. operational satellite nuclear detonation sensors in support of both treaty monitoring and military missions.
- Advance US capability for seismic and radionuclide detection of nuclear tests.



## Missions



- **Safeguards**
- **Warhead Verification**
- **Emergency Response**
  - Find it
  - Characterize it
- **Nonproliferation**
- **Nuclear Weapon Stockpile Stewardship**
- **Nuclear Power Plants**

## An Age-Old Challenge – Unresolved

### Spent Fuel Assay

- determine the elemental Pu mass in spent fuel assemblies
- detect the absence of Pu mass (detect diversions)

There currently exists no safeguards technology capable of independently or directly verifying the plutonium content in spent fuel

## The Challenge

### Safeguards need to

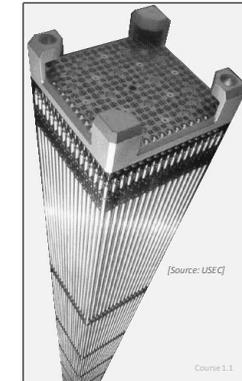
- recovery from a loss of continuity of knowledge
- verify Pu input accountability value at (re)processing or repository facility
- shipper/receiver difference resolution

### Current method infers plutonium content using

- computer codes (that rely on operator declarations)
- indirect measurements (of total neutrons from curium and gross gamma-rays from fission products)

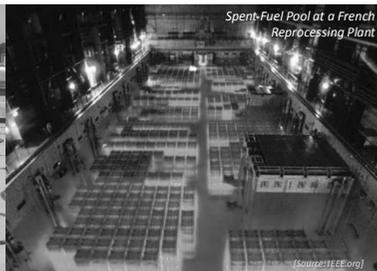
### Constraints

- very complex source terms that vary with fuel type, burn-up, and cooling time
- intense sources of background radiation small margin of error
- average spent fuel bundle: ~5 kg of Pu IAEA significant quantity: 8 kg



## The Risk

- Pu is an inevitable by-product of nuclear power for the uranium fuel cycle
- spent nuclear fuel discharged from a power reactor contains approximately 1 percent plutonium
- approximately 250,000 tons of stored spent nuclear fuel contains 90 percent of the world's plutonium produced by the civilian nuclear fuel cycle



## Fissile Material and Significant Quantity

- Fissile Material
  - Capable of sustaining chain reactions of nuclear fission
  - Capable of undergoing fission by capturing thermal neutrons
  - U-233, U-235, Pu-239, Pu-241
- Special Nuclear Material
  - Plutonium, U-233, Highly Enriched Uranium (> 20%, HEU)
  - Strategic Special Nuclear Material (SSNM): formula quantity
- Significant Quantity (regulated by IAEA)
  - Amount required to manufacture formula quantity
  - 8 kgs U-233
  - 8 kgs plutonium
  - 25 kgs U-235



### U.S., Russian Military Stocks of Highly Enriched Uranium (in tonnes)

End of	Total stocks; average 80 percent	Weapon-grade U 235 equivalent
*1994*	2,030	1,750
1995	2,024	1,744
1996	2,012	1,732
1997	1,985	1,706
**1998**	1,974	1,697
Future***	1,356	1,150

\*These central estimates are from Plutonium and Highly Enriched Uranium 1996.  
 \*Uncertainty: about 20 percent.  
 \*Drawdowns in subsequent years reflect amounts of HEU blended down to LEU by Russia and the United States.  
 \*\* As of July 1998.  
 \*\*\* Future estimates reflect the U.S. government commitment to withdraw highly enriched uranium (HEU) from its military inventory as well as Russia's commitment to blend down HEU (assumed to be all weapon-grade uranium) into low-enriched uranium. The two countries have committed to lowering their stocks by about 547 tonnes of weapon-grade uranium equivalent (or about 618 tonnes of HEU of various levels of enrichment). The schedule is uncertain, but disposal is expected to occur before 2020.

## Global Stocks of Fissile

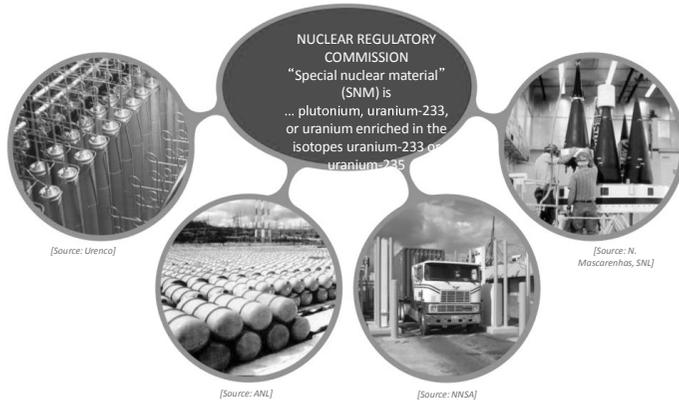
Global stocks (in metric tons)*			
Category	Plutonium	HEU	Total
Civil stocks (rounded)	1,700	175	1,875
Power and research reactor programs	1,595	50	
Declared excess	107	125 (U.S. only)	
Military stocks	155	1,725	1,880
Primary	155	1,250	
Naval and other	--	175	
Russian HEU declared excess	--	300	
Total	1,855	1,900	3,755

**Fissile material: Stockpiles still growing** by David Albright and Kimberly Kramer November/December 2004 pp. 14-16 (vol. 60, no. 6) 2004 Bulletin of the Atomic Scientists

## Keeping Track of Radioactive Materials

- **Safeguards**
  - Make sure it is where it should be
  - Know how much we have
- **Safety**
  - Keep from spreading to unwanted places
  - Prevent unnecessary exposure of personnel
  - Prevent criticality

## On the Hunt for Special Nuclear Material



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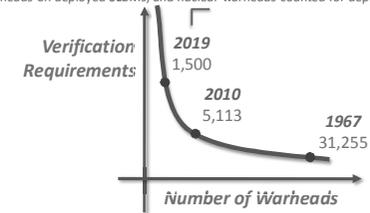
## Arms Control Dynamic

### How low can you go?

new START requires negotiation of the next treaty

real verification will require invasive inspections and technology

deployed ICBMs, warheads on deployed SLBMs, and nuclear warheads counted for deployed heavy bombers

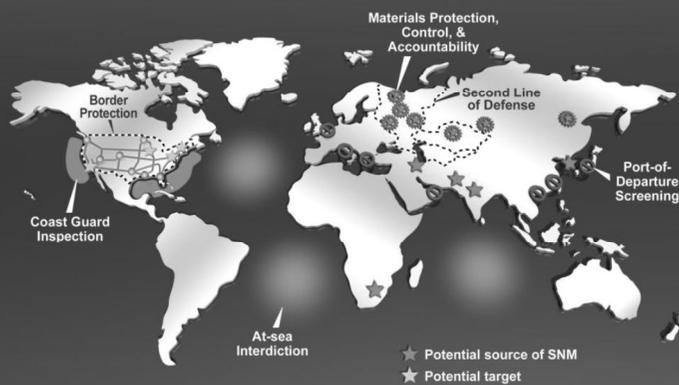


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## U.S. & International Response

### A Global Interdiction System



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## Tomorrow's Challenges

### The road to zero

"... [t]he United States must work to put in place a comprehensive international nuclear-control regime that goes well beyond the present nonproliferation regime's accounting and monitoring of nuclear materials. It must include all fissile materials and provide an airtight verification system to enable the world to move from thousands of nuclear weapons to hundreds, to tens, and ultimately to zero."

from "The Logic of Zero," *Foreign Affairs*, 2008

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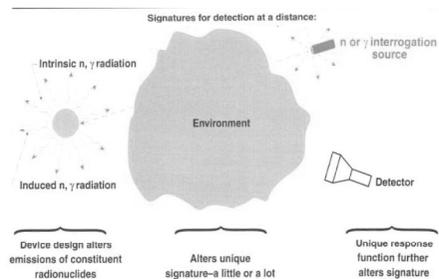
## Detection of Gamma-ray Radiation

- $\gamma$ -rays are produced when atomic nuclei release excess energy after decay reactions
  - Number of  $\gamma$ -rays of each energy released per decay is called branching ratio
  - A single isotope can produce  $\gamma$ -rays at many energies, each with its own branching ratio
- Many  $\beta$ -emitters also emit  $\gamma$ -rays (there are no “pure”  $\gamma$  emitters)
- $\gamma$ -rays are produced at discrete energies
  - Energy remains constant until partial or full interaction
  - Different isotopes produce  $\gamma$ -rays of different energies (known as a signature – can be used to identify emitting isotope)
  - Energy usually referred to in keV

## Passive and Active Detection

- Passive approach is **non-invasive**, requires **relatively inexpensive** equipment, but may not be effective for small amounts of material or under conditions of heavy shielding.
- Active interrogation with neutrons or high energy photons can reveal materials that may be undetectable with passive methods, but requires equipment that is much more **complex, expensive**, and difficult to maintain. Also may involve issues of **nuclear explosive** and **personnel safety**.

## Passive vs. Active Detection



- Passive radiation measurements exploit the radiation signatures provided by the natural radioactivity of fissile material
- Active techniques apply an external radiation source to the object under inspection to stimulate an additional radiation signature

## Passive Detection

- Relies on detection of radiations emitted spontaneously by most nuclear materials .
- Low intensities of gamma rays and neutrons emitted by interesting amounts of material dictate detectors large surface areas operated in pulse mode with high detection efficiencies .
- Self-shielding effects can be strong, even in absence of external shielding

## Passive Detection (Continued)

- The signal falls off as  $1/r^2$  but worsens with
  - Background interference
  - Shielding/intervening materials attenuate and alter the signal
- In descending ease of detection
  - Fixed detector, stationary source (long count, constant BG)
  - Fixed detector, moving source (short count, constant BG)
  - Moving detector, moving source (long count, changing BG)
  - Moving detector, stationary source (short count, changing BG)
- Detection **highly** dependent on device design

## Detection of Gamma-ray Radiation

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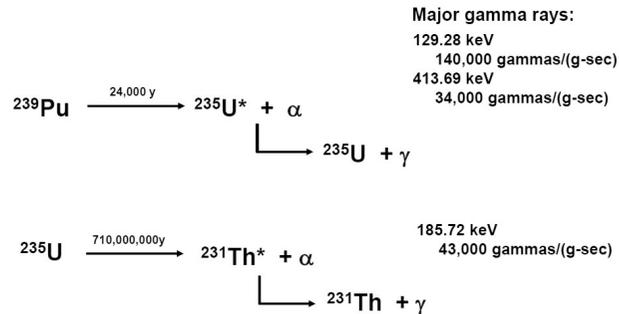
## Considerations in Passive Detection

- Gamma ray energies can be used to identify specific isotopes and/or to discriminate against background radiations -- requires spectroscopic operation of detector with good energy resolution .
- Detection of neutrons significantly above the low background level is almost always an important indicator. Measurement of neutron energy is less important than for gamma rays.

## Uranium and Plutonium

- Emit gamma radiation
- Emit alpha radiation
- Emit infra-red (heat) radiation
  - From  $\alpha$  emission into the matrix
- Emit **singles neutrons**
  - from ( $\alpha$ ,n) matrix interactions and delayed neutrons
- Emit **coincidence neutrons**
  - fission neutrons (from 0 to 8 in a burst)
- Are **fissionable**
  - prompt fission neutrons, delayed neutrons, delayed gammas

## Major SNM Properties



## Useful Radiation Signatures

Isotope	Technique	Signature	Intensity
${}^{235}\text{U}$	passive gamma	186-keV gamma	43,000 $\gamma$ /s-g
	active neutron	1-MeV neutrons	---
$\text{UO}_2$	passive neutron	2-MeV neutrons	0.03 n/g-s <sup>a</sup>
$\text{UO}_2\text{F}_2$	passive neutron	1-MeV neutrons	2.0 n/g-s <sup>a</sup>
$\text{UF}_6$	passive neutron	2-MeV neutrons	5.8 n/g-s <sup>a</sup>
${}^{238}\text{U}$	Passive gamma	1001-keV gamma	100 $\gamma$ /s-g
$\text{Pu}$	passive heat	infrared	---
${}^{239}\text{Pu}$	passive gamma	414-keV gamma	34,000 $\gamma$ /s-g
		375-keV gamma	36,000 $\gamma$ /s-g
		129-keV gamma	140,000 $\gamma$ /s-g
${}^{240}\text{Pu}$	passive neutron	1-MeV neutrons	1000 n/g-s
$\text{PuO}_2$	passive neutron	1-MeV neutrons	120 n/g-s <sup>b</sup>
$\text{PuF}_6$	passive neutron	1-MeV neutrons	7300 n/g-s <sup>b</sup>

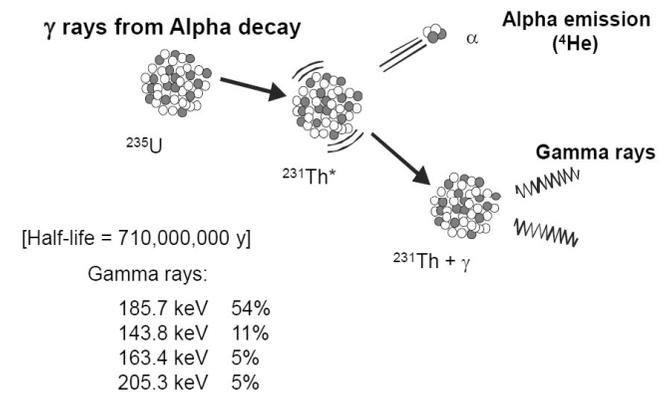
<sup>a</sup> High-enriched uranium with 1%  ${}^{234}\text{U}$

<sup>b</sup> Low-burnup plutonium, with 0.03%  ${}^{238}\text{Pu}$ ,  
6.5%  ${}^{240}\text{Pu}$ , 92.5%  ${}^{239}\text{Pu}$

## Passive Gamma Ray Detection Plutonium

- Plutonium and associated isotopes provide multiple gamma ray lines with energies up to about 650 keV.
- Specific energies and yields are highly sensitive to isotopic mix .
- The higher energy gamma rays can penetrate significant thicknesses of overlying materials .
- Energy resolution better than that from currently available scintillators needed to measure isotopic ratios.

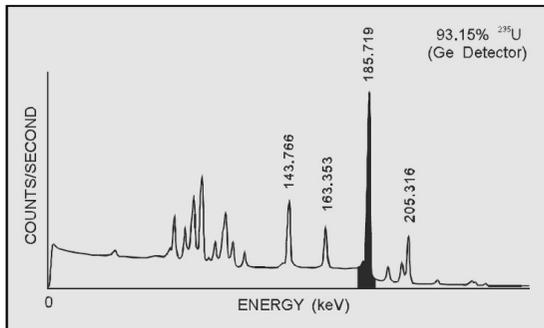
## Gamma-ray Signatures from ${}^{235}\text{U}$



## Passive Gamma Ray Detection Uranium U-235:

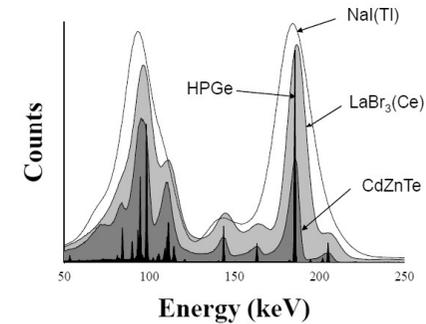
- Weakly penetrating 186 keV gamma ray easily shielded.
- U-238: 1001 keV gamma ray.
- U-232: Highly penetrating 2614 keV gamma ray (from daughter TI-208).
  - Usually (but not always) present in HEU at levels of about 100 ppt.
  - Potential interference from natural thorium that leads to same daughter decay.

### Prominent U-235 Gamma Ray Energies



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### Spectral Resolution of Commercial Detectors (93% Enriched Uranium Photon Spectrum)

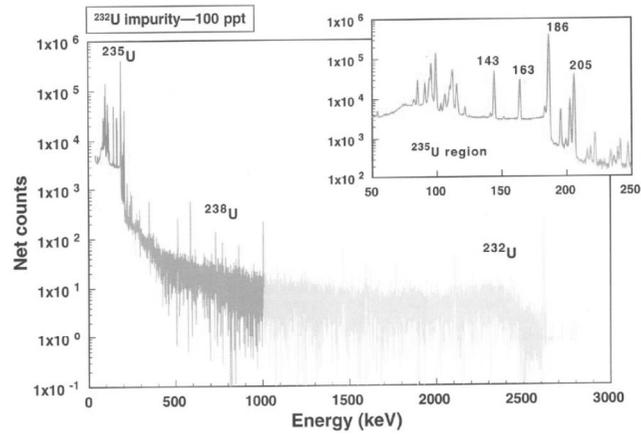


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### Complications in HEU Detection

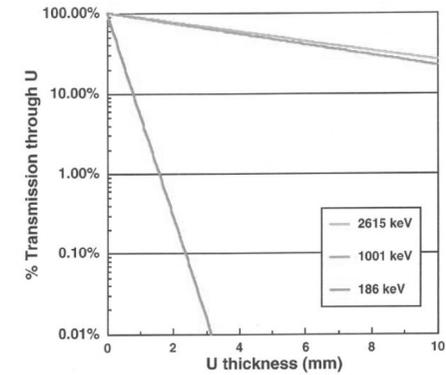
- Passive radiations from fissile material
  - Neutrons
    - Plenty to measure from Pu ( $10^3$  n/g·s)
    - Too few to measure from HEU ( $10^3$  n/kg·s from U-238)
  - Gamma rays
    - Present and strongly penetrating from Pu
    - Plenty but weakly penetrating from HEU
- Unlike plutonium, uranium is a ubiquitous presence in BG radiation that makes weak signals
- Nuclear weapons and components are relatively large, dense, and inhomogeneous: signal highly dependent on device design
- This combination of difficulties makes passive detection of HEU a task ranging from fairly straightforward to impossible.

## High resolution spectrum of oralloy



## Self-shielding Effect

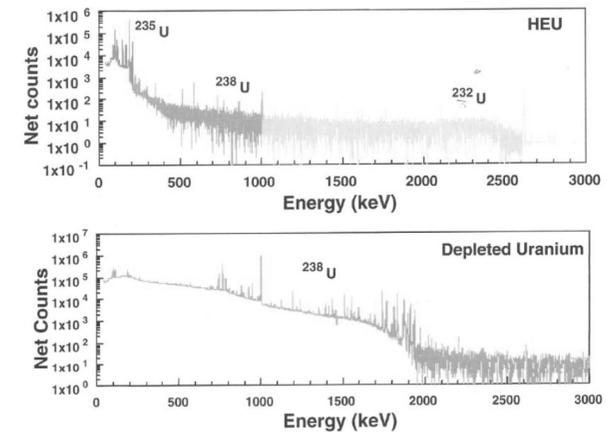
186 keV line: easier to shield than 1001 keV line



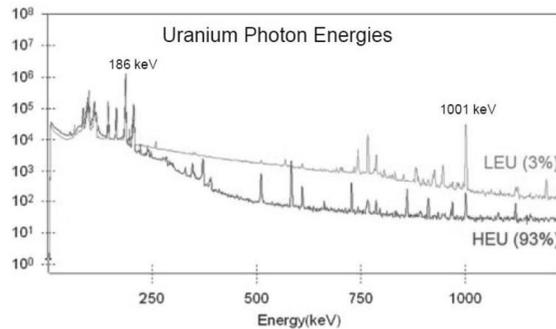
## Principal gamma-ray lines (HEU)

- 186 keV ( $^{235}\text{U}$ )
  - Most intense line emitted by  $^{235}\text{U}$ .
  - Weakly penetrating
  - Identifies presence of  $^{235}\text{U}$
- 1001 keV ( $^{234\text{m}}\text{Pa}$ , granddaughter of  $^{238}\text{U}$ )
  - Most intense line associated with  $^{238}\text{U}$
  - Strongly penetrating
  - Uniquely identifies presence of  $^{238}\text{U}$
  - By itself an insufficient indicator of presence of HEU
- 2615 keV ( $^{208}\text{Tl}$  decay descendent of  $^{232}\text{U}$  trace impurity)
  - Strongly penetrating
  - May infer presence of HEU

## Relative Peak Heights: HEU vs. DU



## LEU vs. HEU

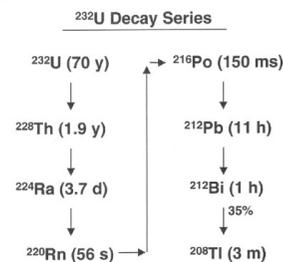


## Potential Use of $^{232}\text{U}$ as HEU Indicator?

- Use of  $^{232}\text{U}$  as an indicator of the presence of U.S. HEU may be feasible
  - It's possible that mere presence of  $^{232}\text{U}$  means HEU in some foreign material
  - Need greater knowledge of the amount (and variation) of  $^{232}\text{U}$  to the amount of  $^{235}\text{U}$  in the material being measured
    - Maybe problematic for some foreign materials
  - Background  $^{208}\text{Tl}$  may obscure the signal from deeply imbedded HEU

## $^{232}\text{U}$ and the 2615-keV Gamma Ray

- $^{232}\text{U}$  is not a naturally occurring uranium isotope
  - Produced in nuclear reactors
  - Appears as a contaminant in the U isotope separation cascade due to reprocessing reactor fuel
  - It will appear preferentially in the low-mass fraction of the isotope separation
- The 2615-keV gamma ray is emitted from the decay of  $^{208}\text{Tl}$
- BG  $^{232}\text{Th}$  also decays to  $^{208}\text{Tl}$ , masking the  $^{232}\text{U}$  signal



## Passive Detection of HEU

- Presence/Absence of HEU
  - Presence of 186-keV line indicates presence of  $^{235}\text{U}$ , but not necessarily presence of HEU
  - Absence of 186-keV line does not indicate absence of HEU
  - Ratio of 186- and 1001-keV lines may indicate presence of HEU
  - Presence of  $^{232}\text{U}$  may serve as a surrogate indicator of the presence of HEU
  - Absence of  $^{232}\text{U}$  is not an indicator of the absence of HEU
  - Ultimately, concentration of individual lines is self-defeating: to maximize S/N we must analyze the full spectrum
- For passive detection of shielded material, these approaches may not be effective in some cases.

## Active Interrogation with Neutrons

- Source can be low-voltage accelerator producing d-d (3 MeV) or d-t (14 MeV) neutrons, or higher voltage accelerator using reactions to produce neutrons of variable energy.
- Some information on enrichment can be obtained if neutron energy is variable (e.g., 1 MeV threshold for fission in U-238).
- Standard technique is to use pulsed source, then look for induced fission neutrons as a function of time.
- Can yield information on multiplicity (related to mass of material being interrogated.)

## Active Interrogation with High Energy Photons

- Source can be electron accelerator (typically around 10 MeV) producing a yield of pulsed bremsstrahlung photons.
  - Average energy of photons can be varied by changing accelerator voltage.
- These penetrating photons produce neutrons in the object under interrogation through (n,gamma) and fission reactions.
- Again look at time dependence of induced neutrons and gamma rays to identify materials.
- Changes with accelerator voltage may provide some differentiation between possibilities.

## Active Detection of HEU: The Promise

- Active techniques are promising because externally applied radiation can penetrate to deeply imbedded HEU
  - Current techniques successfully determine the mass and enrichment of HEU in small, standard, homogeneous samples
  - These conditions do not hold for nuclear explosives
- Examples of active techniques that can be used:
  - Nuclear Material Identification System (NMIS)
  - Threshold neutron-induced fission
  - Neutron-induced differential die-away
  - Bremsstrahlung-induced photofission

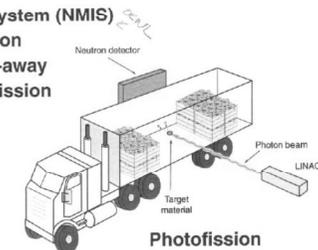
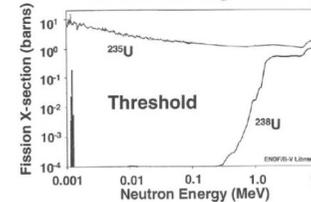


Fig. 3/24/60 — 16

## Active Detection of HEU: Drawbacks

- Active methods have historically been avoided
  - Booby traps—the radiation switch
  - Exposure of live warheads to incident radiation
  - Divergent interrogation sources
    - Inspection personnel and detector exposure problems
    - Stowaways
    - Impractical for long-distance detection  $1/r^5$
    - Not all interrogation sources are divergent
  - Possible need for dedicated facility
- Nevertheless, for shielded material, this is the only game in town

## To Improve HEU Detection

<u>Improved method</u>	<u>Payoff</u>
<ul style="list-style-type: none"> <li>• <b>Passive detection</b> <ul style="list-style-type: none"> <li>– Gamma-ray imaging</li> <li>– Networks of detectors</li> </ul> </li> <li>– Improved signature recognition</li> </ul>	<ul style="list-style-type: none"> <li>– Improved signal-to-noise</li> <li>– Provides effective increase in detector size</li> <li>– Improves signal-to-noise</li> </ul>
<ul style="list-style-type: none"> <li>• <b>Active detection</b> <ul style="list-style-type: none"> <li>– Gamma-ray interrogation</li> <li>– Neutron interrogation</li> </ul> </li> </ul>	<ul style="list-style-type: none"> <li>– Highly penetrating through low-Z material</li> <li>– Highly penetrating through High-Z material</li> </ul>
<ul style="list-style-type: none"> <li>• <b>Improved detector materials</b></li> </ul>	<ul style="list-style-type: none"> <li>– Operational improvements; better energy resolution, lower power, smaller size</li> </ul>

## Holdup Measurement

- Holdup: The residual amount of SNM remaining in a processing facility after bulk materials have been cleaned out.
- Why to measure?
  - **Criticality Safety:**
    - Unanticipated concentrations
  - **Health Hazard:**
    - Radiation and contamination control
  - **Safeguards:**
    - Theft, diversion, material accounting
  - **Economics:**
    - Intrinsic value of SNM

## Causes of Holdup

- Gradual sedimentation or settling in regions of poor circulation (in cracks, pores, corners, etc.) or low flow rate
- Solid or liquid product formation or precipitation resulting from inadvertent chemical reactions
- Internal features
- Electrostatic deposition and buildup of charged particulates
- Splashing, bubbling, or cracking resulting from unregulated chemical reactions

The magnitude of this material will depend on the plant layout, frequency of process upsets, maintenance and cleanout procedures, and throughput

## Typical Regions of High Holdup

- **Pipes and Ducts**
  - Elbows
  - Junctions
  - Seams
  - Changes in diameter
  - Regions of low or stagnant flow
- **Air Filters**
  - Impedance to air flow
  - Particulate barrier
- **Heavy Equipment**
  - Impeller blades
  - Furnace entrances
  - Storage tanks
  - Dissolver trays



## Holdup and Inventory Differences

- Holdup ⇒ Hidden Inventory
  - ⇒ Material Unaccounted For (MUF)
  - ⇒ Diversion ?
  - ⇒ Safeguards Problem!
- Facility operators:  
“Holdup is a fact of life!”
- Regulators:  
“Where is the material?”
- Media:  
“How many bombs are missing?”

## Typical SNM Amounts as Holdup

Gloveboxes	0-50 g
Gloveboxes (after destructive cleaning)	2 g/m <sup>2</sup>
Grinders	1-100 g
V-blenders	1-50 g
Glovebox prefilters	2-100 g
Final filters	10-100 g
Equipment interiors (after routine cleaning)	10-50 g/m <sup>2</sup>
Pipes (after destructive cleaning)	0.3 g/m
Ducts (no cleaning)	1-100 g/m
Glass columns	1 g
Annular tanks	1-10 g
Raschig-ring-filled tanks (after rinsing)	1-500 g
Dissolver trays	10-500 g
Small calciners	5-50 g
Furnaces	50-500 g
Furnace trays	1-10 g
Incinerators	1000s g
Concrete spill basins	1000s g

## Detector Categorization

- Active vs. passive operation.
- Pulse, current, and integrating modes.
- Simple counters vs. spectroscopic systems .
- Omni-directional and imaging detectors

## Common Gamma Ray Detectors

- Geiger tubes and ion chambers .
- Scintillation detectors
  - NaI(Tl) and CsI(Tl)
  - Recently-developed cerium-activated materials .
- Semiconductor detectors
  - - Germanium (requires cooling)
  - -Room-temperature materials (CdTe, CZT, ....)

## Common Neutron Detectors

- Pressurized He-3 tubes
- B- or Li-containing scintillators (bulk or fiber)
- B, Li, or Gd conversion foils used with conventional charged particle detectors
- Fission chambers

## Issues in Gamma Ray Detection

- Detection efficiency .
- Energy resolution .
- Time response .
- Available sizes .
- Cooling requirements .
- Cost

## Issues in Neutron Detection

- Detection efficiency (how "black to thermals" is black enough?) .
- Gamma ray discrimination is critical (background suppression) .
- Scalability to large areas .
- Energy resolution is secondary but could be useful

## Some Observations on Gas-filled Detectors

- Geiger tubes and proportional tubes have unacceptably low gamma-ray detection efficiency for applications of interest here.
- However, proportional tubes with He-3 filling are common neutron detectors with good efficiency.
- High-pressure xenon is an attractive detection medium for gamma rays, most commonly utilized as pulse-mode ion chamber.
  - Could be scaled to large detector area.

## Some Observations on Scintillation Detectors

- NaI(Tl) and CsI(Tl) have been most popular in applications for 5 decades and still are predominant .
- New scintillation materials began a renaissance a decade ago, leading to surprising results for energy resolution and time resolution.
  - Excellent light yield and linearity of response.
  - Some of the newer materials (usually cerium activated) have recently shown results much closer to currently-available RT semiconductor detectors in terms of energy resolution and available sizes.
  - They may now be at their statistical limit, but RT semiconductors are far from their statistical limit.

## Some Observations on Ge

- Mature technology, superb energy resolution near statistical limit .
- Up to 1 kg detectors commercially-available but costly .
- Will always require cooling (small bandgap) .
  - Battery-powered coolers to temporarily replace LN2 under development

## Room- Temperature Semiconductors

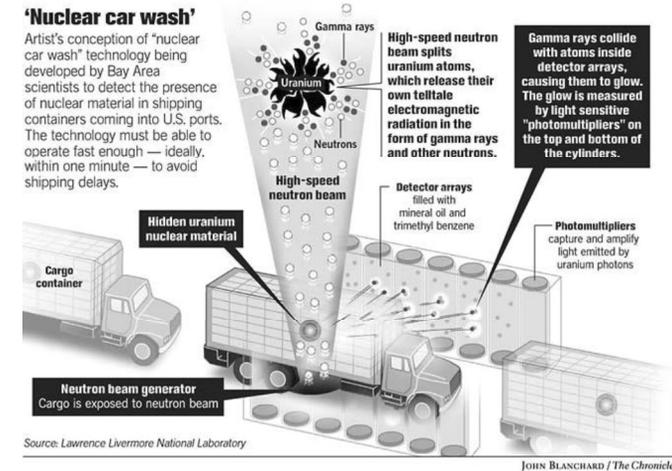
- Some candidates (CdTe, Hgl2) have been investigated for several decades . Largest current activity on Cd-Zn-Te .
- Limitations on size and uniformity in current crystal growth techniques .
- Poor hole mobilities often necessitate more elaborate electronic readout than for Ge or Si
  - Alternative materials (e.g, Aluminum antimonide) under current investigation

## Imaging Detection Systems

- Potential benefits of source localization and background reduction .
- Some approaches:
  - - Conventional shielding and collimation
  - - Coded aperture imaging
  - - Compton imaging

## Some Interesting New Detector Technologies

- Cryogenic micro-calorimeters and superconducting tunnel diodes.
- Silicon avalanche photodiodes .
- Silicon drift photodiodes .
- Microstructure gas detectors and GEMs
- Microchannel plates for neutron detection .
- ASICs for multichannel detector readout .
- Digital pulse processing



## Major Trends in RDM R&D (Hardware)

- Detection Materials
  - Scintillators: SrI2, CeBr2 (gammas), Elpasolites, Plastics (PSD)
  - Semiconductors: Compounds (TlBr, HgI2) — RT Operation
  - Unconventional Detectors: Superconductors (TES)
- Detection Methods
  - Position Sensing combined with Imaging Methods
  - Neutron Detection (Fast and Slow) and Neutrino Detection
  - Electronics: Front-end (ASIC), Readouts (SSPM)
- Application
  - Medical Application: New RDM Methods in Application
  - Homeland Security: SNM Detection, Nuclear Safeguards
  - Scientific Research: Dark Matter Search, Particle Tracks

**MIT** Material Identification & Object Imaging using Nuclear Resonance Fluorescence

**Materials:** Explosives • Nuclear materials • Drugs • Chemicals • Toxic compounds  
**Containers:** Suitcases • Trucks • Ongoing containers • Boxes of wood, iron, aluminum, etc.  
**Advantages:** Very penetrating • Scalable for small and large objects • Unambiguous identification of atomic species and quantitative mass determination • Non-destructive

**The Science**

- Like excited state atoms, characteristic nuclear states. These states are very narrow in energy, and rarely more than a fraction of an eV wide. They can be excited by the absorption of photons of the correct energy.
- When the excited state decays, the characteristic photons are released into all directions with respect to the incident beam, leading to unique photon energies of resonance fluorescence.
- The released photons can be detected by high-resolution solid-state or electron spectrometers.
- Each nuclear species in the beam can be identified by the unique energy spectrum of the released photons.
- The probabilities or cross sections for resonantly absorbing photons are very large. However, penetration is very deep because of Cooper scattering from thermal motion.
- The angular distribution is a gentle function of angle, and depends on the angular momentum of the ground state,  $J_g$ , and of the excited state,  $J_e$ . Coherent phenomena is not critical.
- Excitation by electron background of 500 eV gives reduced photon energy below 0.5 MeV. Thus, there is minimal background from Compton scattering.
- Photon beam is continuously distributed up to the electron energy (non-relativistic). Photons are available at nuclear energies.
- The object is imaged by the collimated photon beam and the collection area of the detector.
- Selection scattering materials can be used to examine transmitted spectra by narrow line absorption. In this manner, the absorption of characteristic photon energies can be measured. This method of material detection is complementary to the detection of released photons.

**Realization**

- Basic science is well established.
- System hardware and software engineering required.
- Activity pending requests for prototype development.

Photon Power Spectrum from the Tungsten Radiator

Material Identification & Object Imaging

High Resolution Deconvolution Spectrometers (SiPMs & CCDs)

Examples of Nuclear & Fluorescent Photon Lines

Resonance of  $E_e, T = 81 \text{ eV}$

$$\sigma = 4\pi \frac{[E_e - E_g]^2 + (\Gamma/2)^2}{\lambda^2 \Gamma^2 G}$$

$$G = \frac{2J_g + 1}{2J_e + 1} \quad \lambda = \text{photon wavelength}$$

Sample Detector Spectra of Resonance Fluorescence

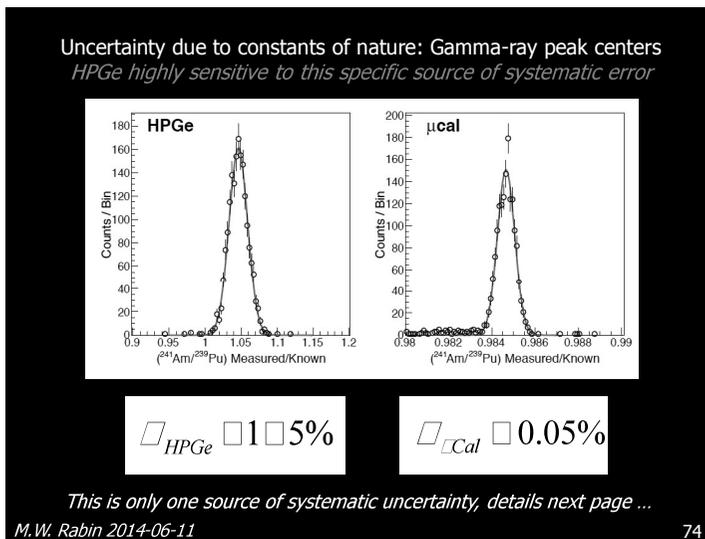
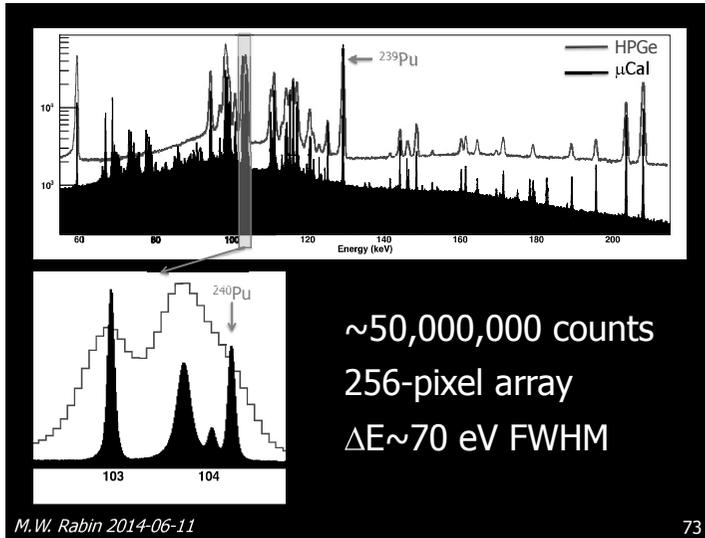
Glass, approximately 1 kg

Nitrogen

Close look at Oxygen lines

Water & alcohol

The techniques illustrated by this material are included in US Patent Numbers 5,422,009 and 5,115,492



## Imaging Technology



- **Gamma - CZT, HPGe, NaI?**
  - Isotope ID, Gamma source localization and geometry
- **Thermal neutron – <sup>3</sup>He and replacements**
  - Location and low resolution image of thermal neutron source (shielded sources)
- **Fast neutron – liquid, plastic, crystal scintillators**
  - Pulse shape discrimination (PSD) – neutron, gamma, etc.
  - Particle identification, low resolution neutron spectroscopy, location of and geometry of fast neutron source
- **Combinations + Contextual Sensing**
- **Muon**
- **Active Neutron Interrogation**
  - Transmission
  - Elastic scatter
  - Tomography – multiple points of view
- **X-Ray**

### Imaging Modes

- Pinhole
- Coded Aperture
- Compton Scatter (> 500 keV)

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## Scenarios



### 1. Locate hot spots in a room or space (~30 meters)

- Possibly high radiation background
- Possibly multiple sources
- Possibly denied access
- Possibly long dwell



ORNL/SNL NCA at SNS

### 2. Characterize the contents of a black box.

- Possibly an immovable black box
- Possibly with restricted access, is in the corner of a room
- Space limitations

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## Gamma Imagers



Germanium Gamma Camera (GGC) by PHDs



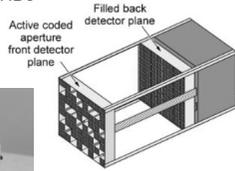
PHDs pinhole aperture



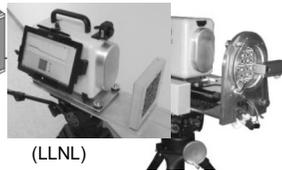
Germanium Gamma-ray Imager (GeGI) by PHDs



Polaris (CZT) by H3D



HEMI by LBNL



Coded aperture (ORNL)

77



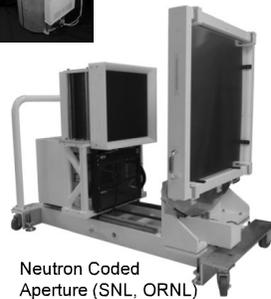
## Neutron Imagers



MINER fast neutron scatter camera by SNL



BNL Thermal Neutron



Neutron Coded Aperture (SNL, ORNL)



High sensitivity directional radiation detection system (rotating) by SNL

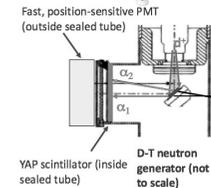
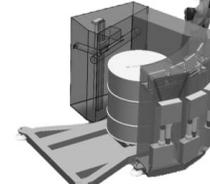
78



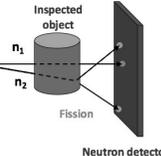
## Active Interrogation Imagers



FNMIS – Fieldable Nuclear Materials Inspection System by ORNL



Fast, position-sensitive PMT (outside sealed tube)  
YAP scintillator (inside sealed tube)



D-T neutron generator (not to scale)  
Neutron detector

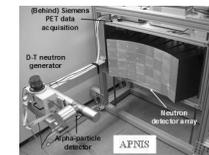
Associated Particle Imaging (API) neutron generators



Thermo API 120  
Optical alpha detector  
5x10<sup>4</sup> n/s



Russian ING-27  
Solid state alpha detector  
1x10<sup>4</sup> n/s



APNIS – Advanced Portable Neutron Imaging System (ORNL)<sub>79</sub>



## What Next?



- **Neutron Generators**
  - Associated Particle Imaging (API) neutron generator availability (alpha detector, small beam spot for good angular resolution, fast timing, higher yield) and field deployable
- **Data management and ease of use**
  - Automated and real-time analysis and results
  - Confidence and uncertainty
  - Information provided to the user (what is actionable)
  - Data transfer
  - File format
- **Stabilize on specific method/information**
- **Performance in the field**
  - Laboratory to field (equipment, measurement degradation, etc.)
- **Communication with the community**
  - Localization versus Imaging
  - Imaging standards

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## Terrorists have redefined the problem

### Secure all U.S. ports including

- Air
- Sea
- Land

### Need to detect for

- Chemical and biological agents
- Special nuclear materials
- Dirty bombs
- Explosives
- Weapons
- Drugs



## At sea ports containers are large, mostly full, and arrive at the rate of ~20 per minute 24/7



~11.6 M maritime cargo containers enter US ports each year\*

\*Jens Baer, McNeill and Jessica Zuckerman, "The Cargo-Screening Clog: Why the Maritime Mandate Needs to Be Re-examined," Background, No. 2357, January 13, 2010.

## Terrorists have redefined the problem

### Secure all U.S. ports including

- Air
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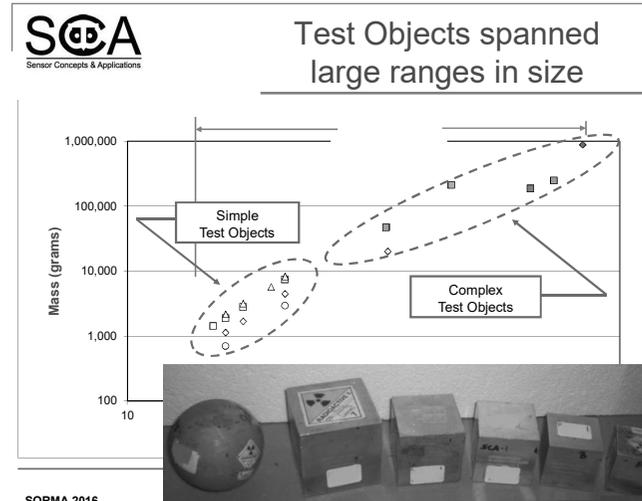
### Need to detect for

- Chemical and biological agents
- Special nuclear materials
- Dirty bombs
- Explosives
- Weapons
- Drugs

### Operational environment

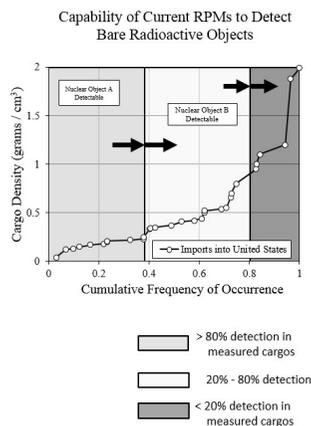
- Negligible impact on commerce
- Time < 1 min/container
- $P_D = 1$  (100%)
- False negatives < 0.05
- $P_{FA} = 10^{-3} < 0.1\%$
- Cargo: Dose < 500 mrem (stowaways)

Define technologies required



## RPMs SNM Detection in US Cargo

- Radiation Portal Monitors (RPMs) are the primary method for initial inspection at US ports
- Their detection performance is well understood and depends on:
  - The threat material,
  - The presence of shielding,
  - The density of surrounding cargo.
- The detection threshold for different SNM threats is mapped to the distribution of inbound cargo.
- For bare objects (no shielding outside of the cargo) the detection depends strongly on the threat type
  - Nuclear object A is detectable in ~38% of cargo containers entering the US
  - Nuclear object B is detectable in ~80% of cargos

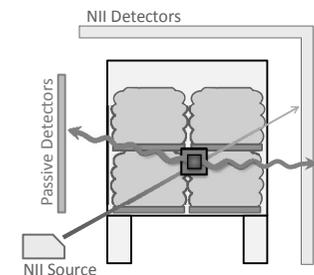


## Passive signatures can be attenuated by both cargo and engineered shielding

**Engineered Shielding:** local material that immediately surrounds the threat attenuating passive signatures (gray).

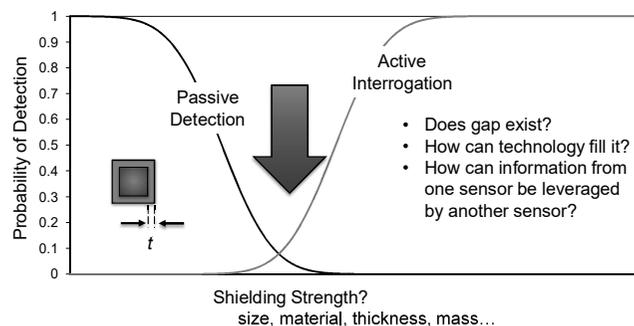
**Cargo Shielding:** distributed material throughout the container that attenuates passive and obscures radiographic signatures. (green)

**Total Shielding:** total attenuation due to cargo and engineered shielding. (green + gray).

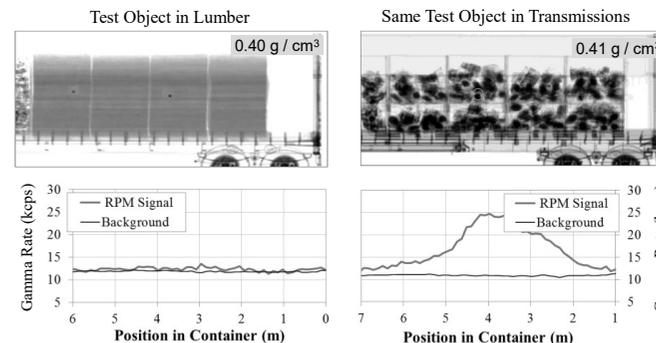


Need a shielding definition that can be applied multiple modalities

## Shielding can reduce passive signatures, but increases visibility to active interrogation

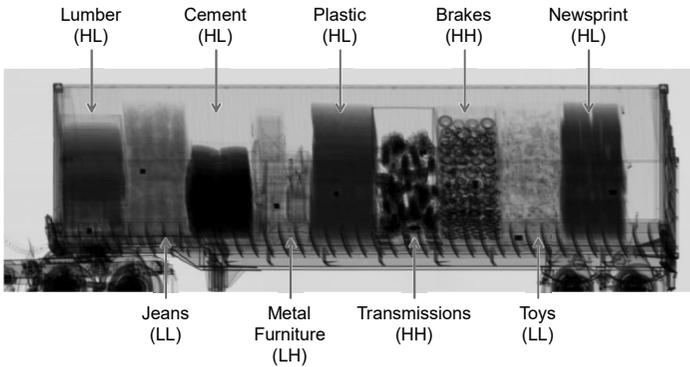


## Passive and Active signatures are strongly affected by cargo complexity and density

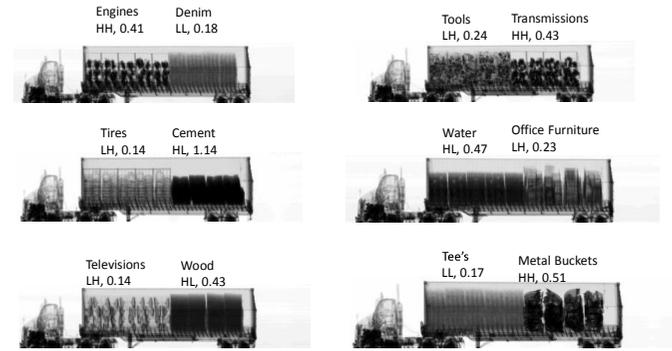


Streaming paths in complex cargo reduce the effective passive shielding

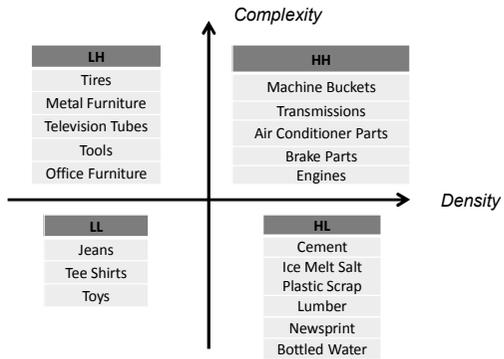
**Systems were tested using 19 cargos representing the range of density and complexity seen in US imports**



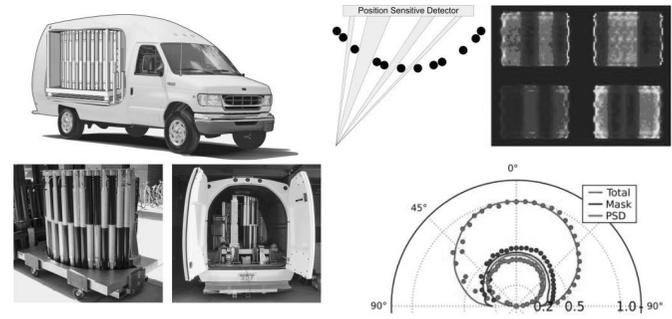
*Standard Cargo*



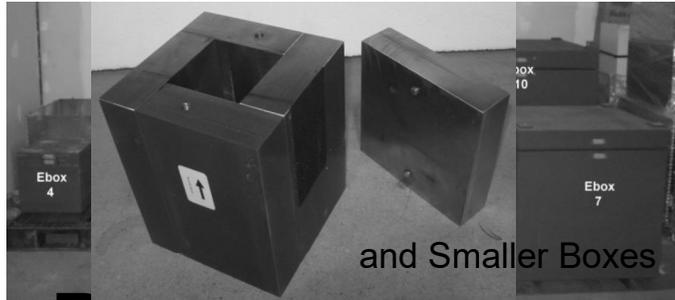
*Representative Cargo Classified on Two Attributes*



SORIS: Stand-off Radiation Imaging System



Engineered shielding was used in combination with test objects



and Smaller Boxes

# Big Boxes ...

Programs focused on Advanced Technologies

		COTS	CAARS/ SNAR	CAARS	SNAR	NRIP
Active Inspection →	Interrogation Signal	X-Rays	X-Rays	Dual Energy X-Rays	X-Rays	Muons
	Method of Analysis	Transmission	Transmission	Transmission	Backscatter Spectrum + Prompt Neutrons	Tomography
	Detection Decision	Manual	Automated	Automated	Automated	Automated
Passive Inspection →	Measured Signal	Gamma & Neutron	—	—	—	Gamma
	Detection Decision	Automated	—	—	—	Automated
Systems Tested *		MSE-1 MSE-2	ASE-1 ASE-2	DE-1 DE-2	BS-1	MT-1

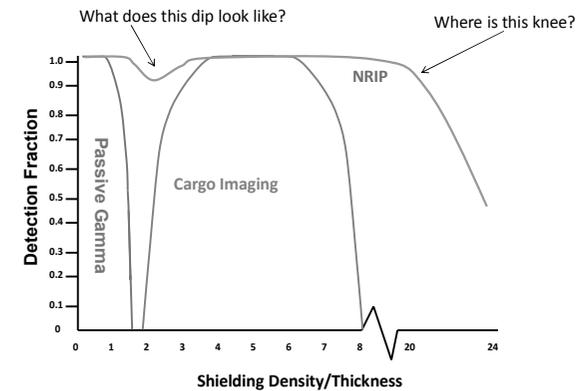
\*Nomenclature:  
MSE-n = Manual Single Energy (x-ray) System Number n  
ASE-n = Automated Single Energy (x-ray) System Number n  
DE-n = Dual Energy (x-ray) System Number n  
BS-n = High Energy Backscatter (x-ray) System Number n  
MT-n = Muon Tomography System Number n

Although details varied, program goals were similar

Primary Inspection Parameter	CAARS	SNAR*	NRIP
Nominal Minimum Threat Object Size	100 cm <sup>3</sup> of High-Z material (Z > 72) behind 10-inches of steel	100 cm <sup>3</sup> of Highly-Enriched Uranium or Weapons Grade Plutonium	Req: 4 kg Special Nuclear Material Goal: 2 kg Special Nuclear Material
Maximum Allowable Fraction of Missed Detections	≤ 10%	Req: < 5% Goal: < 0.1%	Req: < 1% Goal: < 0.1%
Maximum Allowable False Alarm Rate	< 3% of scans with Z < 57	Req: < 5% Goal: < 1%	Req: < 5% Goal: < 1%
Maximum Inspection Time	≤ 1.5 minutes	Req: < 30 minutes Goal: < 30 seconds	Req: < 2 minutes Goal: < 30 seconds

\*Note: Some SNAR Secondary Inspection parameters were applied to Primary Inspection.

## Hypothesis

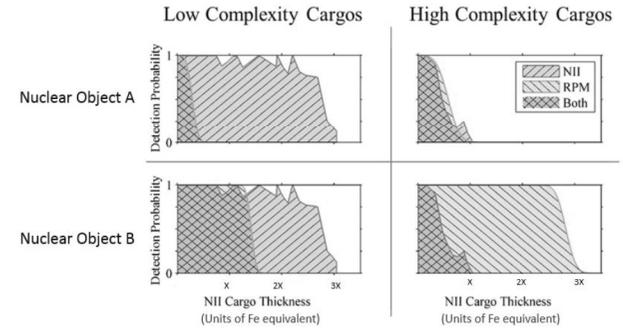


## NII Attributes

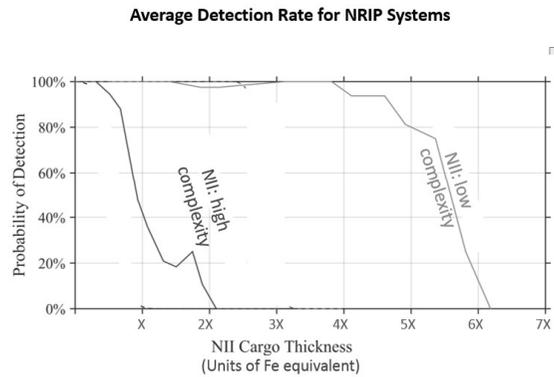
- Well suited for the detection of high-z material (such as SNM)
- Provides significantly more information about container contents, enabling the detection of other contraband.
- Expensive
- Not able to detect radioactive signatures
- Susceptible to false alarms/ false negatives in dense, heterogeneous cargos
- Effective against large portion of end user mission space



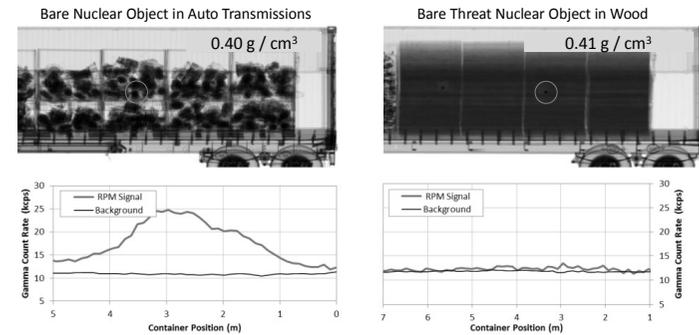
## NII & RPM are Complementary Detection Platforms



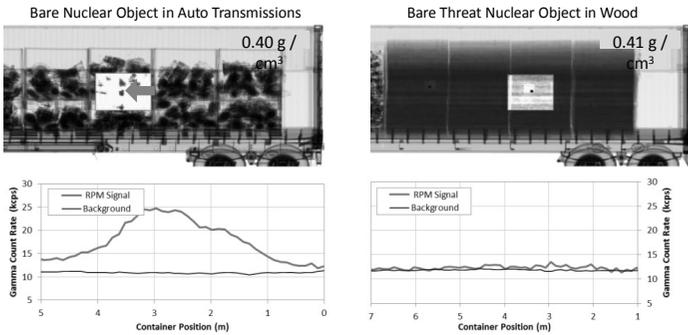
## COTS NII Performance



## NII and RPM Detection performance is cargo-dependent

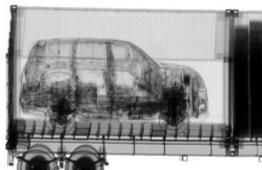
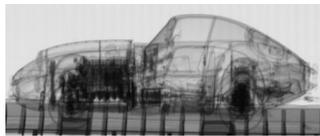


*NII and RPM Detection performance is cargo-dependent*

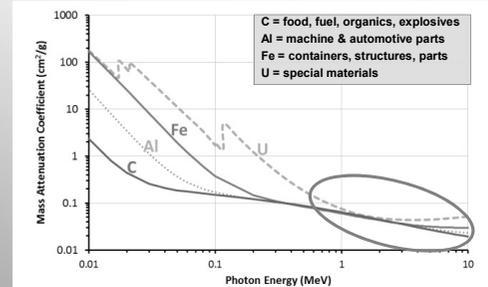


*Information Content: Radiography*

Manifest has HS Code 87 "Vehicles and Parts." Is this a \$10k Nissan SUV or a \$100k classic Jaguar?

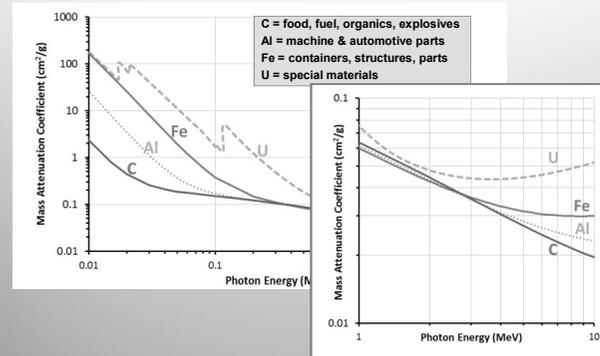


At higher (~1 MeV) energies required to penetrate cargo and vehicles, it is difficult to distinguish different elements/materials



Thus, high-energy x-ray imaging is mainly an anomaly detector

At higher (~1 MeV) energies required to penetrate cargo and vehicles, it is difficult to distinguish different elements/materials



Thus, high-energy x-ray imaging is mainly an anomaly detector

## SAIC/Leidos VACIS II

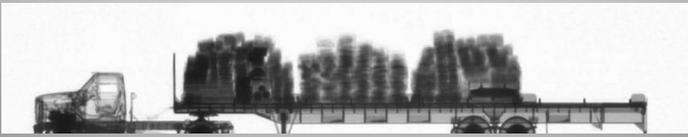
### System configuration



Images from VACIS brochure  
www.leidos.com/products/security

- Sources:
  - $^{137}\text{Cs}$  (0.66 MeV)
  - $^{60}\text{Co}$  (1.2 MeV)
- Detector: NaI
- Source and detector move together on trolleys.
- Circa 1998-present

### Sample Non-Intrusive Inspection (NII) image



## Rapiscan Mobile Eagle 60-DE

### System configuration



Image from Eagle Mobile brochure  
www.rapiscan.com

- Source: Linatron, interlaced 4.5/6 MeV
- Detector:  $\text{CdWO}_4$  array, Si photodiode readout
- Can scan in drive-by or drive-through (stationary) modes.
- Circa 2010-Present

### Sample image



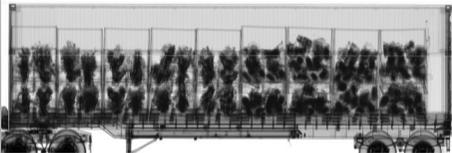
## Smiths Detection HCVP 6030

### System configuration

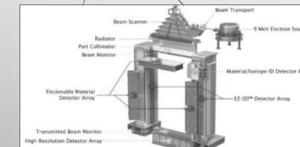


- Source: 6 MeV Linatron
- Detector:  $\text{CdWO}_4$
- Source and detector are fixed, vehicles drive through.
- Circa 2003-present

### Sample Non-Intrusive Inspection (NII) image

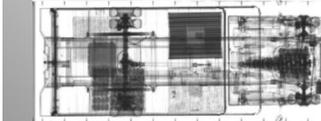


## Passport Systems NRIP Scanner

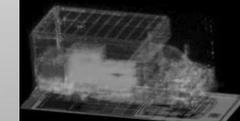


- Source: 9 MeV Rhodotron X-rays collimated to parallel pencil beams.
- Detectors:
  - NaI Array (backscatter)
  - $\text{CdWO}_4$  Array (transmission)
- Three-dimensional volume reconstructed from backscatter signal.
- Prototype as of 2016

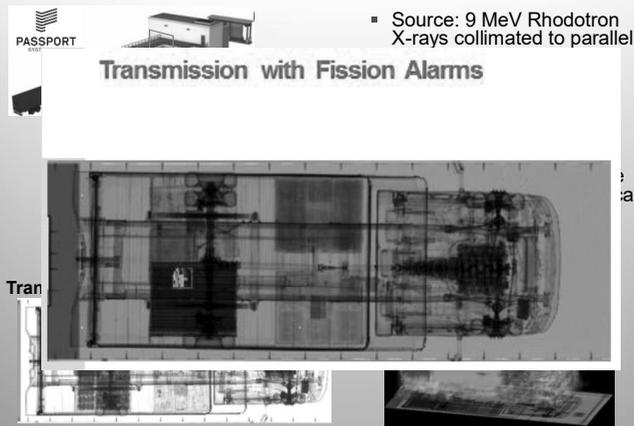
### Transmission Radiograph (top-down)



### EZ-3D™ Reconstruction



# Passport Systems NRIP Scanner



Lawrence Livermore National Laboratory Courtesy of Passport Systems

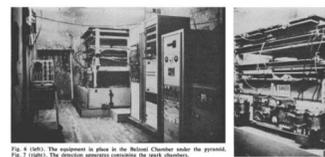
# Stopping cosmic ray radiography



## Search for Hidden Chambers in the Pyramids

The structure of the Second Pyramid of Giza is determined by cosmic-ray absorption.

Luis W. Alvarez, Jared A. Anderson, F. El Behari, James Burkhardt, Ahmed Fahmy, Adin Gheata, Amir Gombel, Viktor Hrabec, Pierre Lavigne, Gerald Lynch, Joseph Miller.



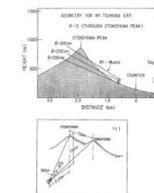
Method of probing inner-structure of geophysical substance with the horizontal cosmic-ray muons and possible application to volcanic eruption prediction

K. Nagamine<sup>a,b,\*</sup>, M. Iwasaki<sup>a</sup>, K. Shimomura<sup>a</sup>, K. Ishida<sup>b</sup>

<sup>a</sup>Muon Science Laboratory, Faculty of Science, University of Tokyo (UTMEL), Hongo, Bunkyo-ku, Tokyo, Japan

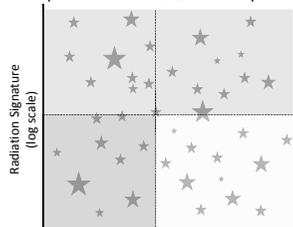
<sup>b</sup>Muon Science Laboratory, The Institute of Physical and Chemical Research (IPAC), Wako, Saitama, Japan

Received 4 July 1994; revised form received 12 September 1994



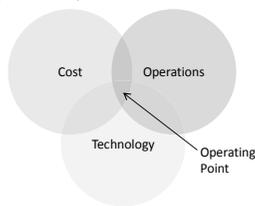
# Better Technology is Needed

Simplified and Notional Detection Space



- Always detected
- Detected if NII scanned
- Never detected

Need better technology that is both operationally viable and affordable

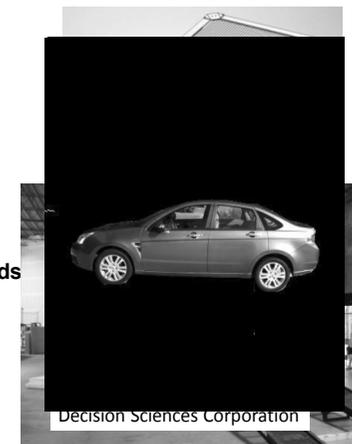


- Detection of all threats through the radiation signature alone is limited by physics
- Improved NII technology
- Greater percentage of cargo be scanned
- Development of other technology to fill the detection space?

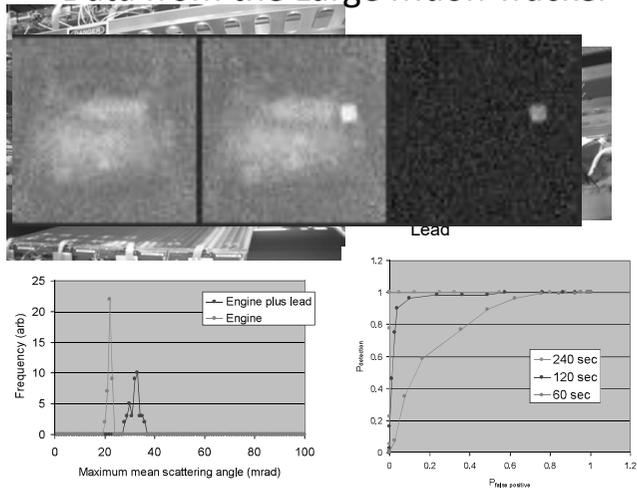


# Scattering cosmic ray radiography

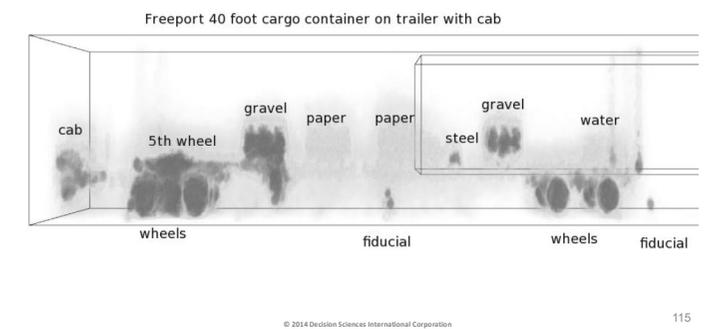
- **Technical approach:**
  - Measure passive radiation
  - Use muons to generate “scattering density” image
    - » Built in momentum measurement
    - » Automatic calibration using flux through empty detector
  - Combine signals to identify threats
- **Advantages over other methods**
  - No radiation
  - Simple technology
  - Inexpensive
  - Can penetrate thick cargos
  - Automatic Identification



## Data from the Large Muon Tracker



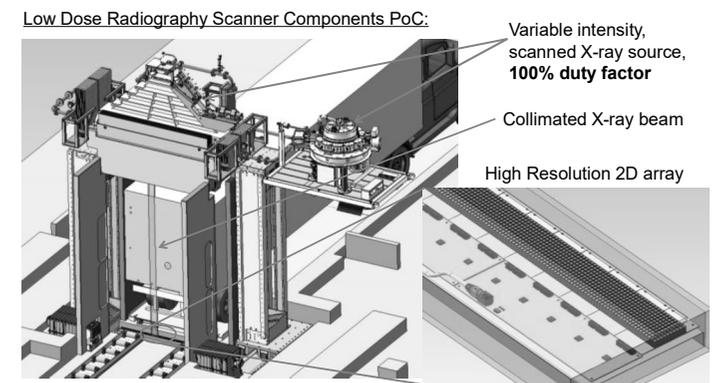
## Freeport



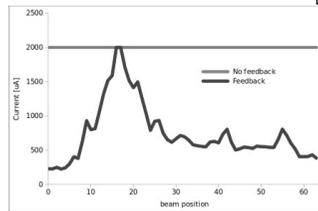
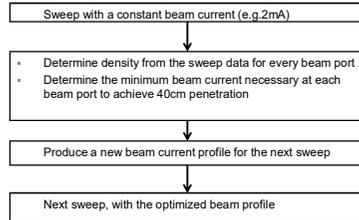
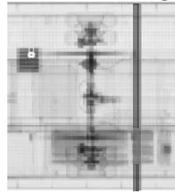
## MMPDS – Freeport, Bahamas



## Program Summary



## Simple Feed-Forward Algorithm



- Necessary development:
  - Fast feedback electronics and precise feedback logic
  - Fast detectors for the imager

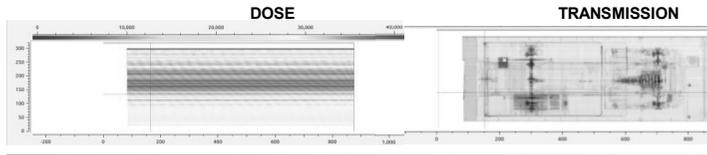


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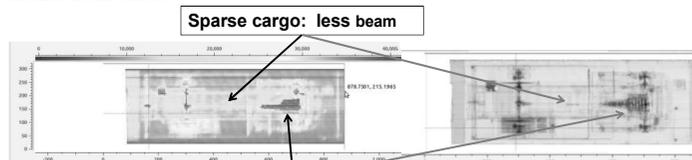
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## Results of the Feed-Forward Algorithm

■ Fixed Beam Current, no feed-forward



■ Variable beam current



Sparse cargo: less beam

Dense cargo: more beam

~ 10x reduction in dose



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## Goal

Develop an active interrogation system that enables SNM detection in a reasonable inspection time and ease in transportability

### Primary Detection Technique

SNM Detection - Differential Die Away Analysis (DDAA)

*Detect prompt fission neutrons after the ENG pulse*

*Most sensitive technique →*

*high thermal neutron fission cross section*

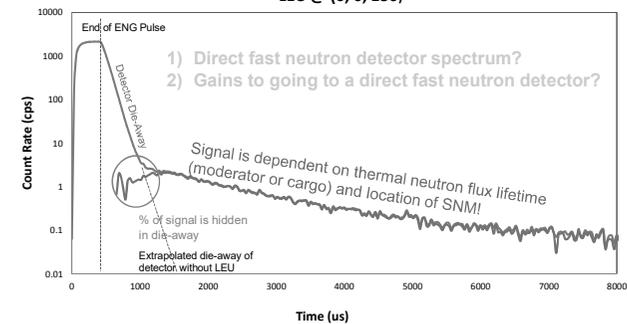
*Delivers minimal dose →*

*low energy (low Q value) of thermal neutrons*



## Differential Die-Away Analysis

Raw & Net DDAA Signal from Moderation-Based Detector  
LEU @ (0, 0, 150)



## System Design

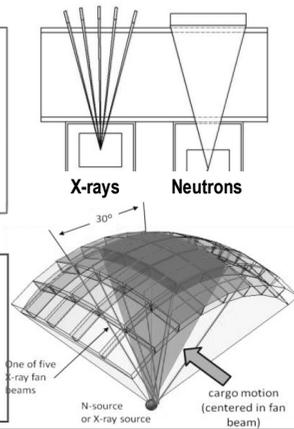
AS&E

### Concepts

- Sought to avoid pitfalls of previous neutron systems
  - Slow, expensive, overly complicated operation
- Tried to use COTS components where available
- Develop hardware that could be manufactured at a reasonable cost
- Wanted 20 cm/s belt speed (standard in airports) with 1 m x 1 m tunnel size

### Design

- Sequential scanning of x-rays and neutrons
- Matched field of views for neutrons and x-rays
  - Use sparse linear arrays for x-rays
- Bottom shooter to reduce neutron shielding weight
- D-D neutron generator (2.5 MeV)
- 170 kV x-ray source



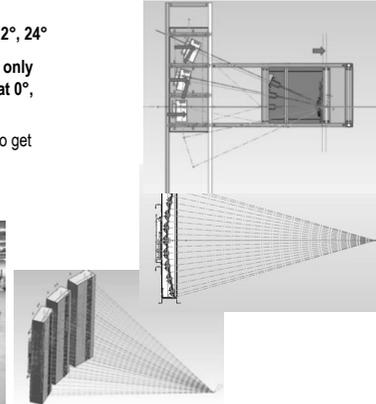
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## X-ray hardware

AS&E

- 5 views arranged at  $-24^\circ$ ,  $-12^\circ$ ,  $0^\circ$ ,  $12^\circ$ ,  $24^\circ$
- To save space and cost for testing, only implemented three physical views at  $0^\circ$ ,  $12^\circ$ , and  $24^\circ$ 
  - Ran cargo through backwards to get other two views
- Used a 140 kV source



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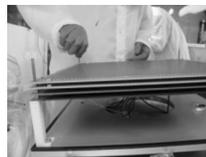
## Neutron hardware and techniques

AS&E



Electro-magnet

- Due to availability we used a DT generator at 14 MeV and a 3 MV Van de Graaff
- Microstructured neutron detector used
  - Single layer used in experiments however multiple layers possible
- Neutrons were in different locations from each other and from the x-rays, requiring careful setup of the experiment
- Performed long counter normalization
- Measured scatter corrections with shadow bars



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