Applied Nuclear Science at Pacific Northwest National Laboratory; The Diverse Work of National Security, International Treaties, and Basic Research

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PNNL
Outline

- Hanford site: a brief history
- Radiological and Chemical Sciences Group
  - Capabilities and Projects
- Comprehensive Nuclear-Test-Ban-Treaty Organization
  - Radioxenon Monitoring
1940’s: Building the Atomic Bomb
The Manhattan Project

University of Chicago
- Theoretical underpinnings of nuclear weapon

Los Alamos
- Weapons design and manufacture

Hanford
- Reactors and Pu production

Oak Ridge
- Isotopic $^{235}\text{U}$ separation
1943: Selection of the Hanford site

- Extremely low population density minimized the need for displacement
- Fast flowing river to cool the production reactors
- Abundant supplies of cheap electricity from large hydroelectric dams
- Remote... to maintain secrecy
- Existing transportation – railroad, barge, and roads
Hanford Site Construction

- 25 million cubic meters of earth moved
- 0.75M cubic meters of concrete poured
- 1.5M concrete blocks and 0.75M cement bricks placed
- 386 miles of roads and 156 miles of rails built
- The first three reactors were constructed (B, D, and F)
- Comparable to 7 major industrial plants.
Termination Winds

During construction the fragile top covering of grass and sagebrush was easily destroyed leaving the sandy and rocky soil exposed to the often fierce desert winds.

New recruits were known to leave after such storms.

This led to the a local song:

*Blow ye winds of Richland*
*Blow ye winds high-o.*
*Blow ye winds of Richland*
*Blow, blow, blow.*

That fearful termination wind,
Can’t stand it anymore;
Each time I sweep
the dust so deep
blows underneath my door.
Hanford’s Primary Mission: Produce Plutonium

1944: The first Hanford reactor to come on line was B reactor.

- Photo: workman loading the core of B reactor with uranium-slug-filled rods
- In the initial hours of operation the reactor began to cool significantly due to $^{135}$Xe poisoning (2,500,000 barn thermal neutron cross-section).
- The reactor operators were able to overcome this problem by loading extra uranium into the reactor core.
- The core capacity had been increased by the reactor engineers unbeknownst to the physicists at the time.

In its first nine months of operation, B reactor produced the Pu used in the Trinity test and the Fatman bomb.
Radiological and Chemical Sciences Group

RC&S Staff Composition

- 80 staff members
- Physics, all kinds but especially nuclear
- Chemistry
  - Radiochemistry / Nuclear Chemistry
  - Analytical Chemistry
  - Physical Chemistry
- Engineering
  - Chemical, Electrical, Mechanical, Nuclear
- Other
  - Materials Science
  - Mathematics
  - Computer Science
Projects & Programs

- Nuclear Material Detection and Characterization
- Process, waste, environment characterization
- Neutrino Physics -- Majorana
- Proliferation detection, especially nuclear test detection
U.S. Customs Support

Instrument US Border Crossings with Radiation Detection Capability

- Characterize threat
  - Gamma and Neutron Detectors
- Apply Commercial of the shelf interdiction technology
- Engineer Installation at USCS point of entry
- Train US Customs Staff
- Work with industry and operations experts for successful technology deployment

High throughput systems

Gamma & neutron detectors
WMD Screening for Cargo Containers

- Need effective yet operationally acceptable sensors to detect threats in inbound cargo containers
  - Assess expected nuclear signatures given realistic cargo, leading to performance specifications
  - Use network of low-cost sensors exploiting long measurement time
Rapidly Deployable Sensor Networks

- Need for effective yet operationally acceptable sensor to detect threats in inbound cargo containers
  - Explore commercial software and hardware capabilities
  - Develop “ad-hoc” network architecture
  - Develop & demonstrate rapid deployment of sensor network
Portable Radionuclide Analysis System

- Rapid quantification of radionuclides in a variety of sample matrices at or near point of collection.
  - Most radioactive decays have a coincidence signature.
  - Portable $\beta-\gamma-\gamma$ coincidence systems
  - Active shielding methods to reduce ambient background effects.
  - Use ROOT as an analysis tools for coincidence signatures
Most special nuclear materials U, Pu, Am emit high fluxes of neutrons.

Field teams need

- A light weight, directional detector for neutron sources at distances of 10 - 100 meters
- A moderator-free detection method for slow neutrons. Air is the moderator.
- Theory: build model describing slow neutron transport to long distances
- Boron shielding and collimation
- Standard $^3\text{He}(n,p)^3\text{H}$ neutron detection.

Nuclear Radiation Sensors
Advanced Land Mine Detection Method

- Need for a low-cost, portable instrument capable of effective and efficient detection of modern buried land mines.

- Use timed neutron detection method to rapidly find mines in arid & semi-arid soil (scan rates > 6 m²/min.)

- Technique shown to work will for real mines (disabled for testing).

- Developed instrument weighs ~5 lbs and costs less than $5K.

- Sensitive to organic and water content of soil, (don't rely on this unit as only technique)
Fiber Optic Development for Sensors

- Specialized optical fibers that are sensitive to neutrons and insensitive to gamma’s
  
  - Developed scintillating glass formulations that meet neutron detection application requirements
  
  - Use the high neutron cross-section of the $^6\text{Li}(n, t, \alpha)$
  
  - Built a fiber draw tower to fabricated specialized optical fibers from scintillating glass
  
  - Individual fibers have low gamma-ray detection efficiency
  
  - Fibers have been deployed in a number of applications
Portable Neutron Spectrometer

- Need field-appropriate instrument that permits measurement / exploitation of neutron energy spectrum.

- Develop, construct, & test field-portable neutron spectrometer.
- Exploit fiber-optic neutron detection medium for novel spectrometer design
- Uses layers of fibers and layers of plastic, and multiplicity counting to discriminate out gamma’s
- Can be used for a variety of purposes: Treaty verification, smuggled weapons search, Environmental remediation…
Pulse Shape Processing

- Use list mode for post data acquisition analysis
  - Energy and time stamps
  - Pulse shape analysis (better resolution)
  - Analysis individual wave forms (dual component)
Dual Optical Component Scintillators

- Need effective $\gamma$–ray rejection or neutron energy determination.
  - Explore $n$ and $\gamma$–detection capability of LiBaF scintillator:
    - a) Determine optical performance,
    - b) Evaluate neutron detection capability,
    - c) Grow larger crystals for practical uses.
  - Can uses same analysis techniques for Phoswich detectors to measure coincident $\beta$–$\gamma$, $\alpha$–$x$–ray, etc signatures.

![Graph showing gamma and neutron pulses](image)

Pacific Northwest National Laboratory
U.S. Department of Energy
Synthetic Gamma-ray Spectra

- The ability to collect reasonable gamma spectrum for a given source, detector type and geometry aids R&D effort

- Developed a set of computational algorithms representing the physics of gamma-ray detection
  - NaI, HpGe, CdZnTe, LaCl₃

- Develop user-friendly software to provide rapid calculation of synthetic “spectra”
  - Runs on Windows

- Validate predicted spectra
- Commercialized & support software “Synth”
Neutrino Research: Majorana Overview

- **GOAL:** Sensitive to effective Majorana $\nu$ mass near 50 meV
- Neutrinoless double $\beta$ decay of $^{76}\text{Ge}$ potentially measured at 2039 keV
- Based on well-known $^{76}\text{Ge}$ detector technology plus:
  - Pulse-shape analysis
  - Detector segmentation
- Requires:
  - Deep underground location
  - 500 kg enriched 86% $^{76}\text{Ge}$
  - Many crystals, segmentation
  - Pulse shape discrimination
  - Time/Spatial Correlation
  - Special low-background materials
Background comparison of several PNNL systems

- Above ground or shallow underground (100 mwe) sites can be quite useful in extending sensitivity, but limited
- Sensitivities deep underground can be 100x better or more

<table>
<thead>
<tr>
<th>Location</th>
<th>Background Detection Limit</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shallow Underground</td>
<td>0.01 0.1</td>
</tr>
<tr>
<td>Well Shielded Surface</td>
<td>0.01 0.1</td>
</tr>
<tr>
<td>Deep Underground</td>
<td>0.0001 0.01</td>
</tr>
</tbody>
</table>

Low-Background Electroformed Copper

- Semiconductor-grade acids
- Glassware-free handling
- Copper sulfate purified by recrystallization
- Baths circulated with continuous microfiltration to remove oxides and precipitates
- Continuous barium scavenge removes radium
- Cover gas in plating tanks reduces oxide formation
- Periodic surface machining during production minimizes dendritic growth

Low-background detector and electroformed cryostat during assembly
CTBT–IMS: what is it?

- Comprehensive Nuclear-Test-Ban-Treaty (CTBT): eliminate nuclear weapons testing
- International Monitoring System (IMS) will be implemented to produce verification data for the CTBT

Four types of networks

- Infrasound, Hydro-acoustic, Seismic
- Radiation: Particulate, Radioxenon
- The Automated Radioxenon Sampler-Analyzer (ARSA) is a radionuclide sampling station developed for application to the IMS
The International Monitoring Sensor Network

IMS RN Network by end of 2003 (of 80 stations)

- 55 stations installed (or 66%)
- 22 stations certified (or 28%)
Production Mechanism

Radionuclides are produced directly from the fission device (fission products) or secondarily (activation products)

- Below ground testing traps most of the fission products.
- Weather carries the escaped radionuclides to the samplers.
- Samplers collection a large amount of air and determine the radioactivity per volume yielding an activity concentration.
- No direct measure of device yield nor device type.
Advantages of Monitoring Radioxenon

- Radioxenon is in mass range with large fission yield
- Several RXe isotopes are produced with convenient half-lives (\(^{131}\text{m} \text{Xe}, \(^{133}\text{m} \text{Xe}, \(^{133} \text{g} \text{Xe}, \text{and} \(^{135} \text{Xe})\)

fission yields for A=130–135

Plot from Kaplan, "Nuclear Physics" (1963)
Advantages of Radioxenon Monitoring II

- Xenon is an inert noble gas, likely to escape even from an underground explosion
- Doesn’t combine with other gases in atmosphere (no chemical effects)
- Easy to extract from air using traps at moderate temperatures (~163K)
- Dominant background: radon, easily removed with proper processing
Automated Radioxenon Sampler/Analyzer

- Separates and concentrates Xe from 87 ppb to 65% in a 6.6 cc volume
- Highest system sensitivity ever produced ~100 µBq/m³
- Only system capable of measuring $^{135}$Xe in the environment
- Fully automated, automatic data transfer and high throughput 48 m³/8 hours - 144 m³/day
General Specifications of the ARSA

- Automatic collection, purification, and analysis of radioactive xenon from the atmosphere
  - Remotely controllable over the internet or modem (or GCI)
  - Self-monitoring software
  - Analysis software included
- Radioxenon concentration measured every 8 hours
- Detection sensitivity for $^{133}\text{Xe}$ better ($<0.1$ mBq/m$^3$) than CTBT IMS specifications
  - Sensitive detection of $^{131m}\text{Xe}$, $^{133m}\text{Xe}$ and $^{135}\text{Xe}$
How does the ARSA Work?

- Collect air, separate, purify and measure xenon, count RXe decays, analyze isotope concentration

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Air Intake

Chiller (-125 C) → Main charcoal trap for xenon

Compressor and traps for H2O, CO2

Xenon purification and measurement → RadioXe Counting

Analysis
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Radio Xenon Gamma/Beta Signatures

131m Xe
11.93 d
163.9 keV

133m Xe
2.19 d
233.2 keV

135m Xe
15.65 m
526.6 keV

131g Xe
(Stable)

133g Xe
346 keV
5.25 d

135g Xe
9.14 h
905 keV

133g Xe
(Stable)

133 Cs
(Stable)

135 Cs
(Stable)

30 keV x-ray in coincidence with 199-keV conversion electron

30 keV x-ray in coincidence with 129-keV conversion electron

81-keV gamma-ray in coincidence with 346-keV beta spectrum

AND

30 keV x-ray in coincidence with beta spectrum plus 45-keV conversion electron

250 keV gamma-ray in coincidence with 910-keV beta spectrum

E_{photon}

E_{beta}
**β–γ Spectrometer: NaI(Tl)**

- **NaI(Tl) detector with 4 wells**
  - NaI(Tl) crystal is nominally 1” thick
  - Two crystals optically separated
  - Each crystal viewed with two 3” PMT’s
  - Size:
    - 18 cm tall
    - 35 cm wide
    - 10 cm thick
Beta-Cell

- Beta-cells made of plastic scintillator, $4\pi$ geometry for $\beta$'s
  - External sources provide QA/QC checks
  - Size:
    - 1 cm right cylinder
    - 5 cm long
    - Wall 1.2 mm thick

Gas (beta) Cell
(6.2 cm$^3$)

Sample Input/Output

Photo-multiplier-tubes
ARSA: Detector Basics

 Entire assembly enclosed in copper cave (5 mm thick) surrounded by 5 cm of lead.
Background Suppression

ARSA: data from EML field test in NYC

- Background is reduced with coincident detection of both the photon and electron

April 2nd, 1997 ARSA Measurement

Counts

Energy (keV)

Gamma-ray singles

- 30 keV
- 81 keV
- 133Xe (32 mBq/m³)
- 135Xe (3.7 mBq/m³)
- 250 keV

Energy (keV)
Two Dimensional Histogram

- 2-D Plot: $\gamma$ vs. $\beta$ pulse height
- Radioxenon isotopes inhabit well-defined regions (plot)

- Beta Resolution clearly shows $^{131m}$Xe (gate on the 30-keV and 80-keV region)

Radioxenon isotopes include:
- $^{214}$Pb
- $^{135}$Xe
- $^{133}$Xe
- $^{133m}$Xe, $^{131m}$Xe, $^{133m}$Xe

Beta Data
Radon Interference

- **β-γ Energy spectrum from a radon spike**
  - 3 α’s, several β-γ coincidences.
  - $^{222}\text{Rn} \rightarrow ^{218}\text{Po} \rightarrow ^{214}\text{Pb} \rightarrow ^{214}\text{Bi} \rightarrow ^{214}\text{Po} \rightarrow ^{210}\text{Pb} \rightarrow ^{210}\text{Bi} \rightarrow \ldots$. 

\[ \text{Gamma Data} \]

High Radon _cell1.png

$^{214}\text{Bi}$ 609-keV
$^{214}\text{Pb}$ 242, 295 352-keV
$^{214}\text{Pb}$ 80-keV
\[^{133}\text{Xe}\] Concentrations in Freiburg Germany

- Measured over 1100 samples,
- High concentrations from nuclear reactors or Hospitals
The Commercial ARSA System

- Commercial ARSA will be available soon
  - Produced by DME in Florida, selling price expected near $650K
  - Taking data in Guang Zhou China since 2001
  - First production units available sometime in 2004
Summary

- Lots more work not shown.
  - Modeling
  - Environmental monitoring
  - Hanford cleanup
  - International policy
  - IAEA work
  - Etc. …..

- Applied Nuclear Research alive and well in the Pacific Northwest.