Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Savannah River National Laboratory™

OPERATED BY SAVANNAH RIVER NUCLEAR SOLUTIONS

We put science to work.™

Stabilization of Residual Contamination with Alternative Materials for Deactivation and Decommissioning of Legacy Nuclear Facilities

Savannah River NUCLEAR SOLUTIONS℠

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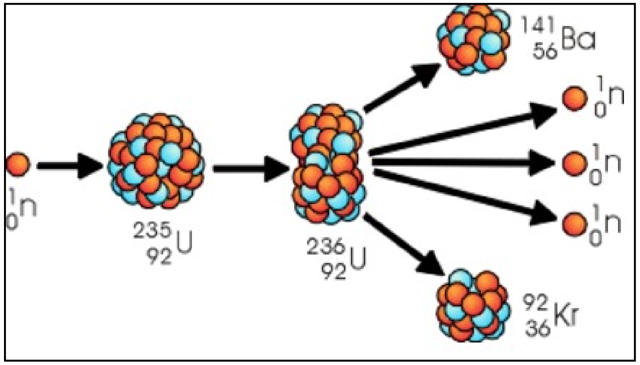
SRNL-STI-2019-00245

The Department of Energy: A Brief History



Dec. 1938

Otto Hahn and Fritz Strassmann discover fission in uranium

Sep. 1, 1939

Feb. 24, 1941

Glenn Seaborg's group at UC Berkeley discovers plutonium

Dec. 7, 1941

The Department of Energy: A Brief History

Jan. 19, 1942

FDR approves production of atomic bomb

Aug. 13, 1942

Uranium isotope separation facilities are built at Oak Ridge, TN.
Plutonium production reactors are built at Hanford, WA.
Weapons laboratory is set up at Los Alamos, NM.


THE BALTIMORE NEWS-POST

EXTRA

10-Star Edition

GERMANY SURRENDERS

London Sets V-E Day For Tomorrow

Truman Waits On Stalin,

May 7, 1945

Uranium bomb, Little Boy is dropped on Hiroshima, Japan.
Plutonium bomb, Fat Man is dropped on Nagasaki, Japan.

Aug. 6 and 9, 1945


JAPANESE SURRENDER; WORLD WAR II ENDS

Pearl Harbor Is Avenged!

AMERICAN POWER OVERWHELMS FOE

The Department of Energy: A Brief History

As the cold war intensifies, new facilities are authorized for the Hanford Site, Oak Ridge, Paducah -KY, Portsmouth and Fernald- OH, Aiken - SC, Idaho Falls - ID, Rock Flats - CO, Amarillo - TX, Livermore -CA, Las Vegas - NV

THE DEMOCRAT
AND LEADER

RUSSIA HAS ATOMIC BOMB!

U. S. Learns of Recent Explosion, President Discloses

Pentagon Shows Excitement, But Officials Mum

Bargaining In Steel Resumed

Russ Possession of Bomb Peace Threat, Congressmen Believe

U. S. Still Ahead of Russians

Details on Blast in USSR Not Revealed; Vishinsky Urges Pact

ALCOA Fights Bettendorf's Adoption Plan

Soviet Union detonates first atomic device

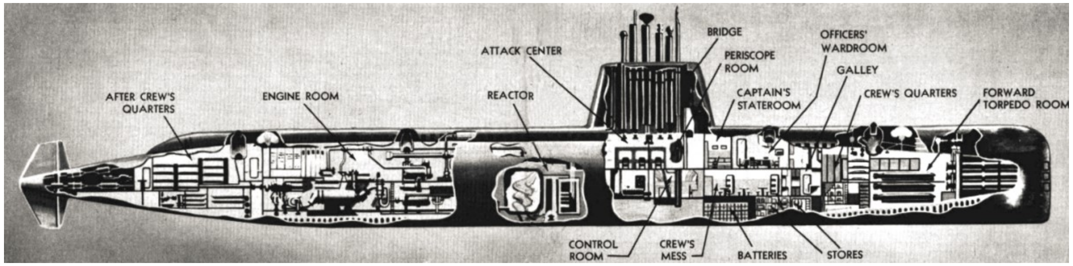
Jan. 1, 1947

Nov. 1947

Aug. 29, 1949

All atomic energy activities are transferred to the newly created Atomic Energy Commission

The Department of Energy: A Brief History



Science paper published on the discovery of the neutrino, a fermion that interacts only via weak subatomic force and gravity, Clyde Cowan and Fred Reines using the flux from P Reactor at SRS

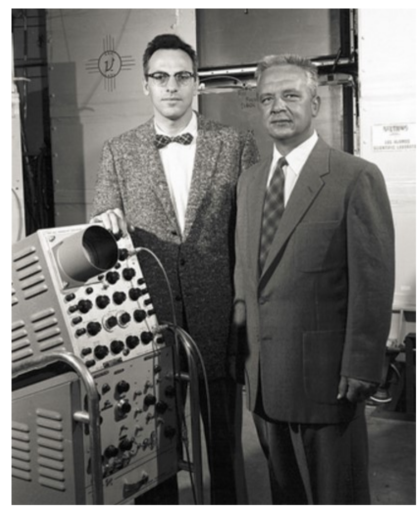
Jan. 24, 1954

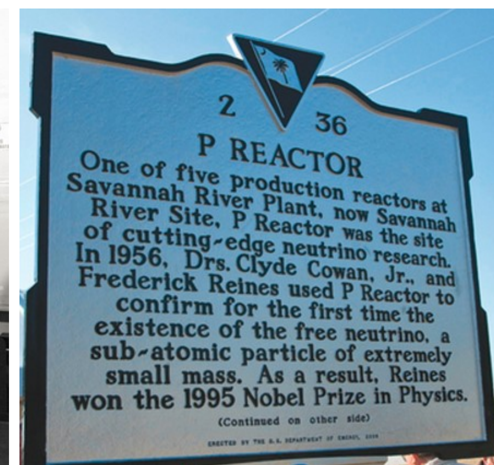
The Navy launches the first nuclear-powered submarine, U.S.S. Nautilus

Dec. 23, 1957

The world's first full-scale nuclear power plant becomes operational at Shippingport, PA

July 20, 1956

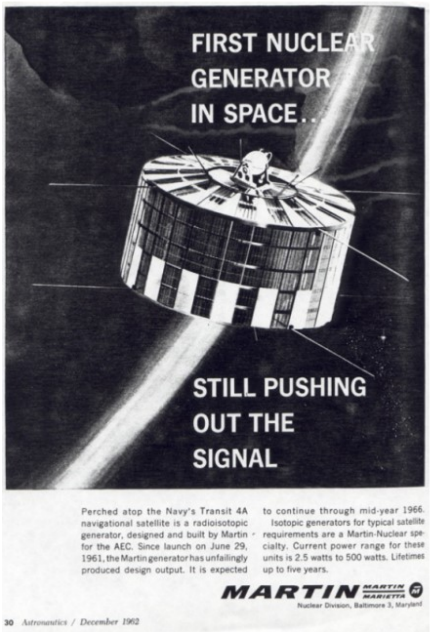




The Department of Energy: A Brief History

June 29, 1961

The first radioisotope thermoelectric generator for space applications is launched on the Navy Transit 4A spacecraft. The used 238-Pu was produced at SRS



June 29, 1961

The Soviet Union breaks a three-year moratorium on nuclear weapons testing. In a 60 day period, the Soviets conduct 50 atmospheric tests with a total yield exceeding all other previous tests.

Sep. 15, 1961

The U.S. resumes nuclear weapons testing with an underground test at the Nevada Test Site.

Mar. 5, 1970

The United States, Great Britain, the Soviet Union, and forty-five other nations sign the Treaty for the Nonproliferation of Nuclear Weapons.

The Department of Energy: A Brief History

President Ford signs the Energy Reorganization Act of 1974, abolishing the Atomic Energy Commission and establishing the Energy Research and Development Administration and the Nuclear Regulatory Commission.

Oct. 11, 1974

President Carter signs the Department of Energy Organization Act. The Federal Energy Administration and Energy Research and Development Administration are abolished.

Aug. 4, 1977

The Department of Energy (DOE) is activated; bringing together a score of entities from a dozen departments and agencies.

Oct. 1, 1977

The Department of Energy: A Brief History

Mar. 28, 1979

A partial meltdown of the core occurs at one of the two reactors at the Three Mile Island nuclear power plant near Harrisburg, Pennsylvania

June 20, 1979

President Carter announces program to increase Nation's use of solar energy, including solar development bank and increased funds for solar energy research and development.

Oct. 8, 1981

The Reagan Administration announces a nuclear energy policy that anticipates the establishment of a facility for the storage of high-level radioactive waste and lifts the ban on commercial reprocessing of nuclear fuel.

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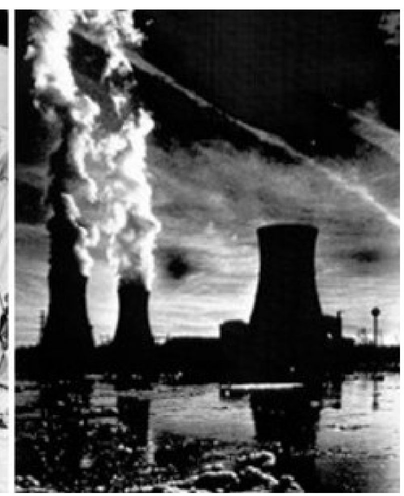
NEW YORK POST FINAL LATEST PRICES

A-PLANT MISHAP LEAKS RADIATION

Contaminated steam escapes in Pa.



The Department of Energy: A Brief History

A major nuclear accident occurs at Chernobyl Reactor #4 near Pripyat, Ukraine in the Soviet Union, spreading radioactive contamination over a large area.

Apr. 26, 1986

President Carter announces program to increase Nation's use of solar energy, including solar development bank and increased funds for solar energy research and development.


"All the News That's Fit to Print"

The New York Times

DECLARING DEATH OF SOVIET UNION, RUSSIA AND 2 REPUBLICS FORM NEW COMMONWEALTH

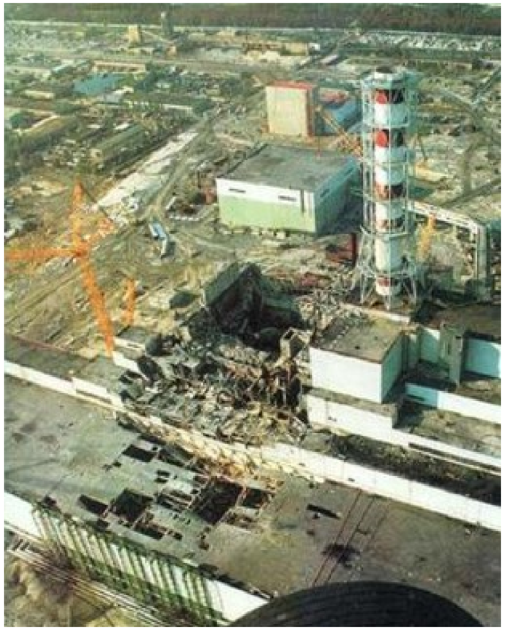
Frantic Moves Came to Light In Days Before Maxwell Died

The New and the Old

TAKE OVER A-ARMS

Newborn Bureaucracy Is Inheriting Functions of Old Authority

West Europeans



June 20, 1979

Nov. 1990

President G. H. Bush declares the end of the Cold War as the Soviet Union collapses.

The Department of Energy: A Brief History

May 10, 1992

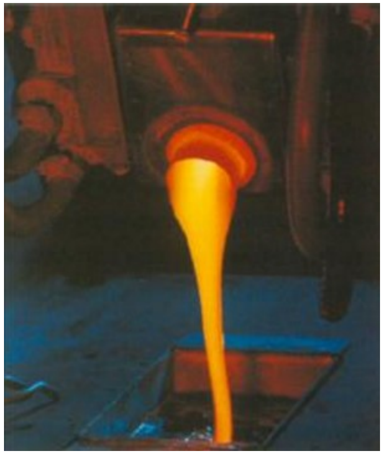
Secretary Watkins testifies before the Senate Armed Services Committee that for the first time since 1945 the United States is not building any nuclear weapons.

Sep. 23, 1992

The United States conducts its last underground nuclear weapons test. Congress imposes a temporary moratorium on nuclear weapons testing

Nov. 23, 1994

The Department announces the completion of a highly classified interagency operation to transfer weapons grade highly enriched uranium out of Kazakhstan to DOE's Y-12 Plant at Oak Ridge, Tennessee.



Mar. 1996

Vitrification of high-level nuclear waste in glass canisters begins at the Defense Waste Processing Facility (DWPF) at the Savannah River Site. The facility is the largest of it's kind in the world.

The Department of Energy: A Brief History

Dec. 19, 1996

DOE unveils a dual-track strategy to dispose of the nation's surplus plutonium.

Apr. 25, 1997

Workers complete drilling of the five-mile long, horseshoe-shaped exploratory tunnel through Yucca Mountain at the proposed high-level nuclear waste repository in Nevada.

June 20, 1997

The Plutonium Uranium Extraction Facility (PUREX), the largest of the Nation's Cold War plutonium processing plants, is deactivated a year ahead of schedule.

Jan. 22, 1998

DOE announces that it will dispose of defense-generated transuranic waste at the Waste Isolation Pilot Plant (WIPP) in southeastern New Mexico.

Dec. 22, 1998

DOE announces that the Savannah River Site will be the location of a plant that will disassemble pits from nuclear weapons and convert the recovered metal to an oxide, beginning a process of destroying rather than creating weapons-grade plutonium.

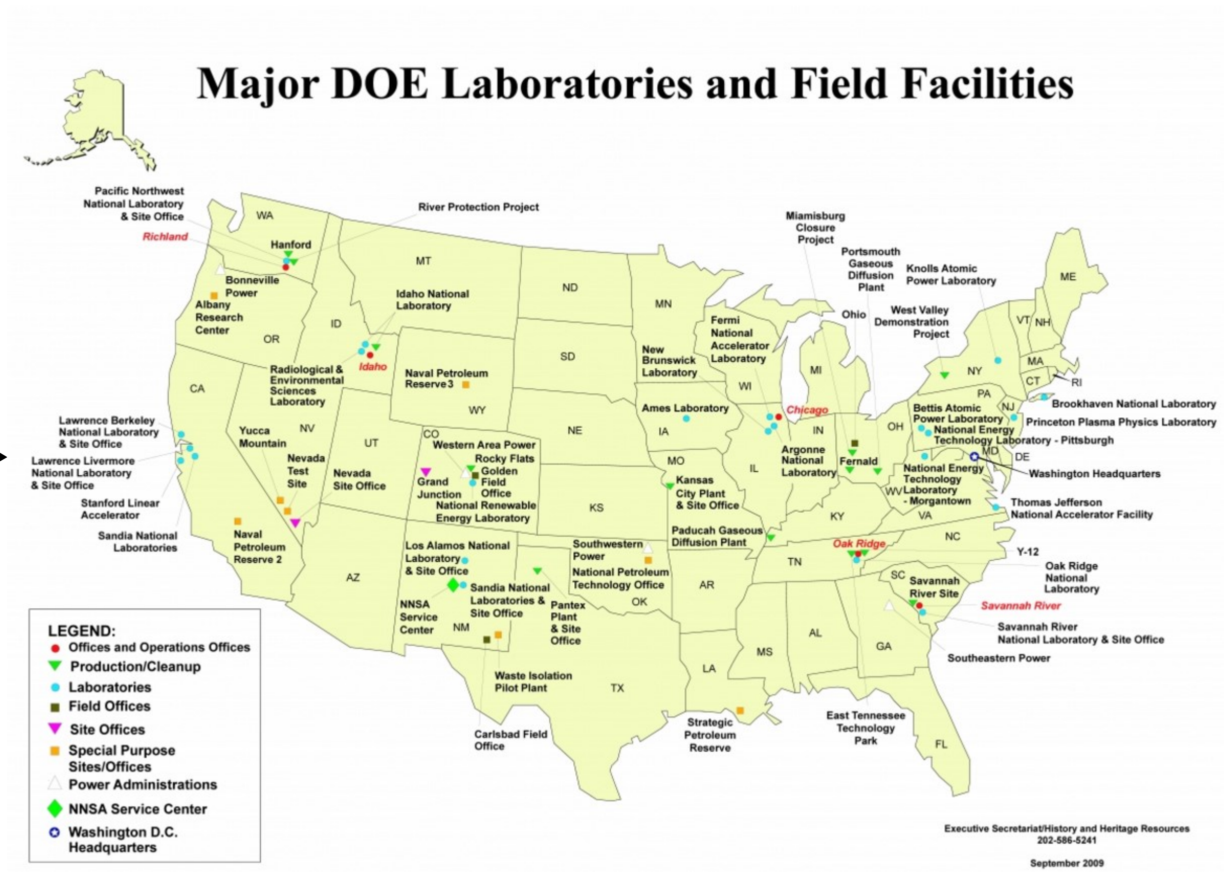
The Department of Energy: A Brief History

DOE activates the National Nuclear Security Administration (NNSA) to carry out the national security responsibilities of the Department of Energy.

Mar. 1, 2000

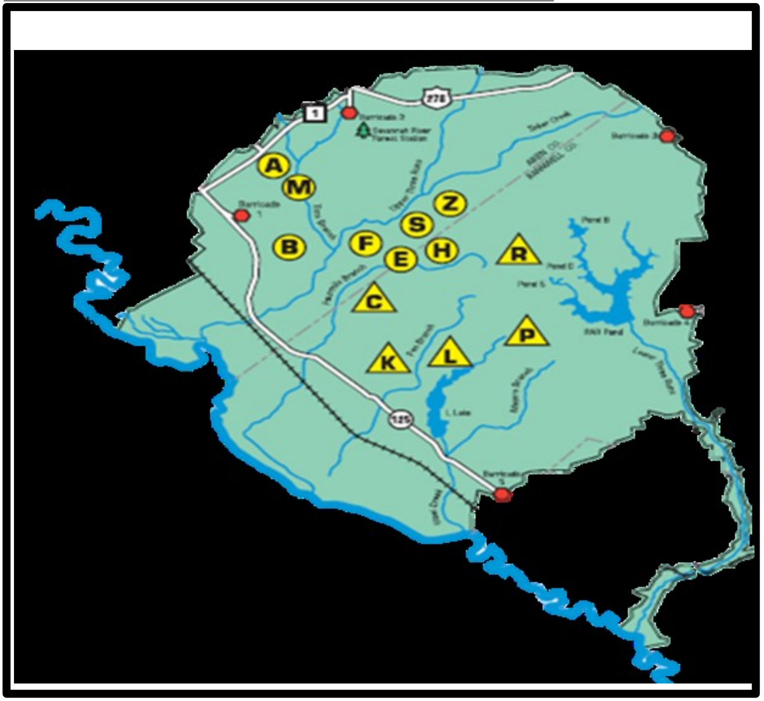
2000 – Present

Work pertaining to waste disposal, clean energy technology, and national security continue within the DOE complex.



DOE National Laboratory System

Savannah River Site Overview



SRS is a key DOE site responsible for environmental stewardship and cleanup, waste management and disposition of nuclear materials.

- **~803 square kilometers**
- **SRS workforce: Approximately 10,000**
  - DOE-SR and DOE-NNSA
  - **Savannah River National Laboratory**
  - **Savannah River Nuclear Solutions (M&O Contractor)**
  - Other contractors include Savannah River Remediation, Centerra SRS, CB&I AREVA MOX Services, Parsons, and the University of Georgia (Savannah River Ecology Laboratory)

History of Savannah River Site

Waste tanks construction - 1957


P Reactor

- **Nuclear materials production history**
  - 5 nuclear materials production reactors
  - 2 separations plants
  - Heavy water extraction plant
  - Nuclear fuel and target fabrication facility
  - Solid and liquid waste disposition processes
- **Environmental legacy**
  - 130 million liters highly contaminated liquid
    - Stored in 47 underground tanks with very limited access
      - Liquid, saltcake, sludge
  - 6 Fuel basins
    - Wide variety of fuels
    - Damaged (corroded) fuel
  - Decommissioned radiological facilities
  - 515 radionuclide or chemically contaminated soil and groundwater waste sites
  - Over 2 x 10^6 m^3 contaminated groundwater

The Savannah River Site Today and the Savannah River National Laboratory

Traditional laboratories and specialized facilities and equipment for both radiological and non-radiological work

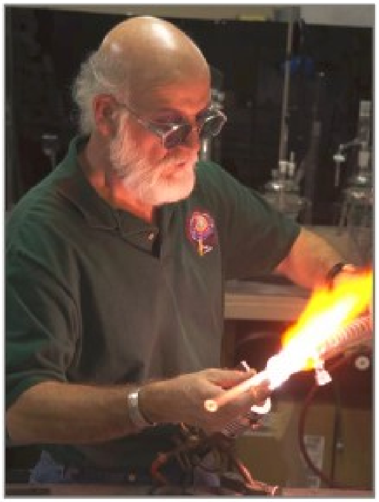
Main Laboratory Building

Shielded Cells for manipulation and testing of highly radioactive material

Atmospheric Technologies for weather and plume tracking and modeling

SRNL technician working with hazardous materials in glovebox

Engineering Development Laboratory for technology scale-up and demonstration



Laboratory for fabrication of specialized glass equipment

The Savannah River Site Today and the Savannah River National Laboratory

- Non-nuclear labs in offsite space
- Other facilities throughout SRS

Grout Development Laboratory for formulation of specialized cementitious materials for use in diverse radiological environments

2 SRNL Analytical Laboratories (F/H and Tritium) provide high quality analytical, radiometric and environmental monitoring data to a range of customers

Laboratory in ACTL

The Savannah River Site Today and the Savannah River National Laboratory

Clean Energy

- Hydrogen Production and Storage
- Nuclear Fuel Cycle R&D
- Renewable Energy Research

National Security

- Nuclear Defense
- Tritium Technology
- Homeland Security
- Nonproliferation
- Nuclear Forensics

Environmental Stewardship

- Waste Treatment
- Materials Stabilization and Disposition
- Remediation and Cleanup
- Assessments and Verification

235-F Risk Reduction Project Drivers & Challenges

DNFSB issued Recommendation 2012-1,
***Savannah River Site Building 235-F Safety* (5/9/12)**

Remove/immobilize Materials At Risk (MAR) from/in Plutonium Fuel Form (PuFF) Facility

235-F Building

Materials At Risk (MAR) *removal is challenging because .....*

- Few penetration into or between confinements (i.e., cells)
- Difficult to manipulate components within the confinements (i.e., cells)
- Maintaining confinement during setup and maintenance

PuFF Shift Operating Base

Incombustible Fixatives - Background

Scope

To develop and characterize a deployable, **fire resistant** radiological contamination fixating platform deployable in non-standard environments (i.e. hot cells, wing cabinets, etc.)

Current Fixative Materials are designed to be applied in "ideal conditions": 70 ºF, 50% humidity & no claimed fire resistance

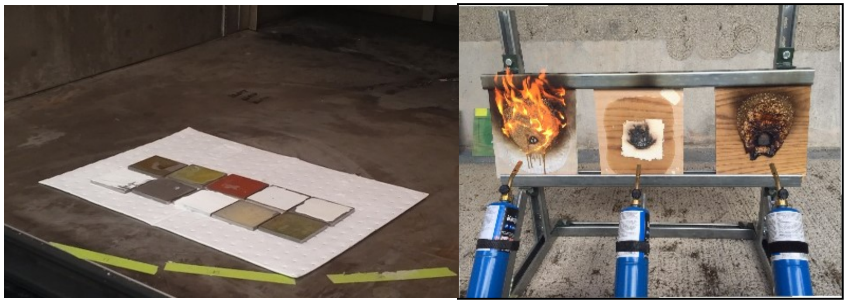
Down Selection

- Environmental (SRNL) – temperature/humidity effects
- Radiological (SRNL) – gamma irradiation 5MRad
- Adhesion (SRNL) –fixative remains adhered to substrate
- Fire (FIU*) – direct flame performance
- Mass Loss (FIU) – fixative performance at discrete temperatures increasing to 800 °F

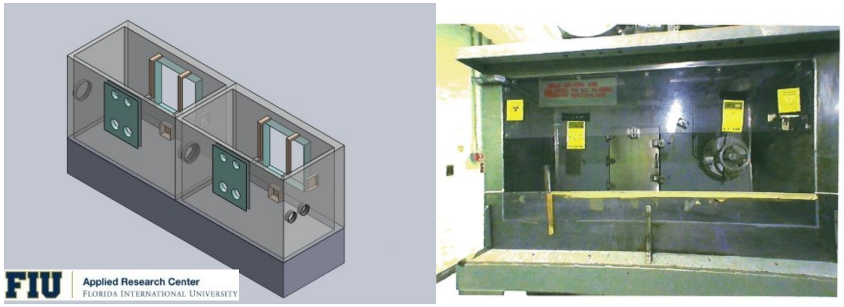
*FIU – Florida International University

Field Testing

- Cold demo completed at FIU (Summer 2017)
- SRNL incorporated cold demo results and prepared Hot Test Plan (Winter 2017)
- SRNL conducted Fixative Cold Demo for operator training at SRS 235-F Mockup (June 2018)
- SRNL conducts Fixative Hot Test/Demo at SRS 235-F PuFF Facility (Ongoing)



SRNL environmental testing (left) and FIU direct flame testing (right)



FIU Cold Test Mockup (left) and Hot Test Stage – Contaminated Wing Cabinet/Entry Hood at SRS 235-F PuFF Facility (right)

Benefits

- Commercial fire resistant materials adapted for radiological application.
- Easily and rapidly applied to vertical and horizontal surfaces
- Stabilization of residual contamination influences facility disposition approach.
- Reduces worker risk levels and technical uncertainty.

Fixative Cold Test – June 2018

Initial spraying was performed exterior to the cell to familiarize operators with sprayer setup, functions, and cleaning. Representative spray areas (~3ft X 3ft) were marked and sprayed multiple times.

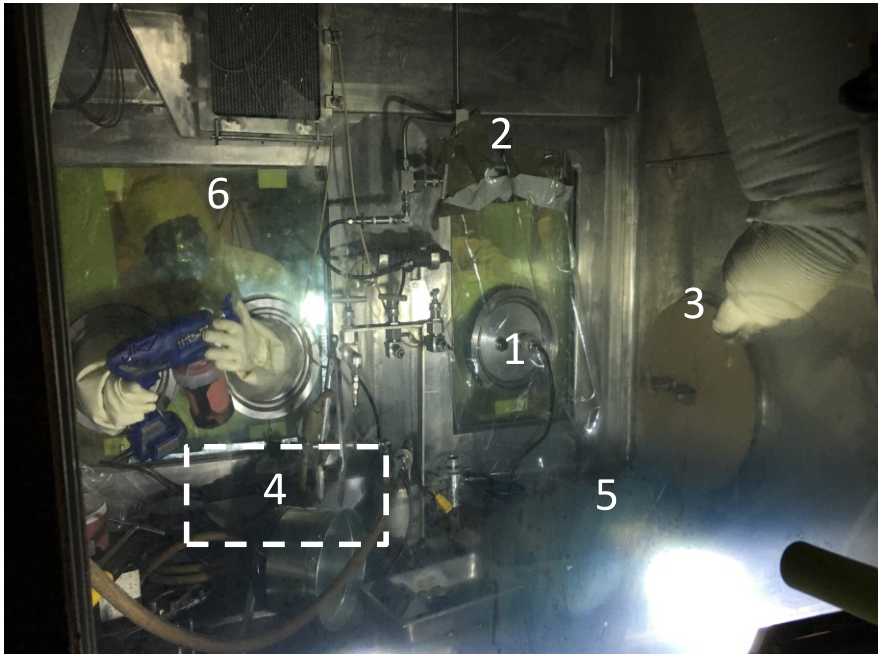
Sprayed and poured/spread was then performed in the 235-F Process Cell Mockup that is the same configuration as Hot Cell 7.

Verification of thickness from cold demo has aided "eye-balling" thickness during hot demo operations.

Lessons Learned – SRNL Cold Demo

- Horizontal vs. vertical spray was easier to control overspray
- If too much FD is applied at one time, sloughing can occur
- Curing in a stagnant, closed system takes significantly longer than when air is circulated
- Thickness gauge can give extraneously low results if backing is not metal
- Total spray time of one quart (one cartridge) is **~1.5 minutes**

Entry Hood and Cell 7 Preparation



- Installation of flex wall (1)
- Run ACE for ~24 hours (2)
- Clear debris inside hood (3)
- Mask square door (4)

- Installation of electrical pass-through (1)
- Shield electrical pass-through from overspray (2)
- Protect Cell 7/8 transfer lock (3)
- Run ACE for ~24 hours (4)
- Clear debris near spraying area (5)
- Bag in sprayer, pre-loaded cartridges, and other necessary equipment (6)

Spray Application in Entry Hood

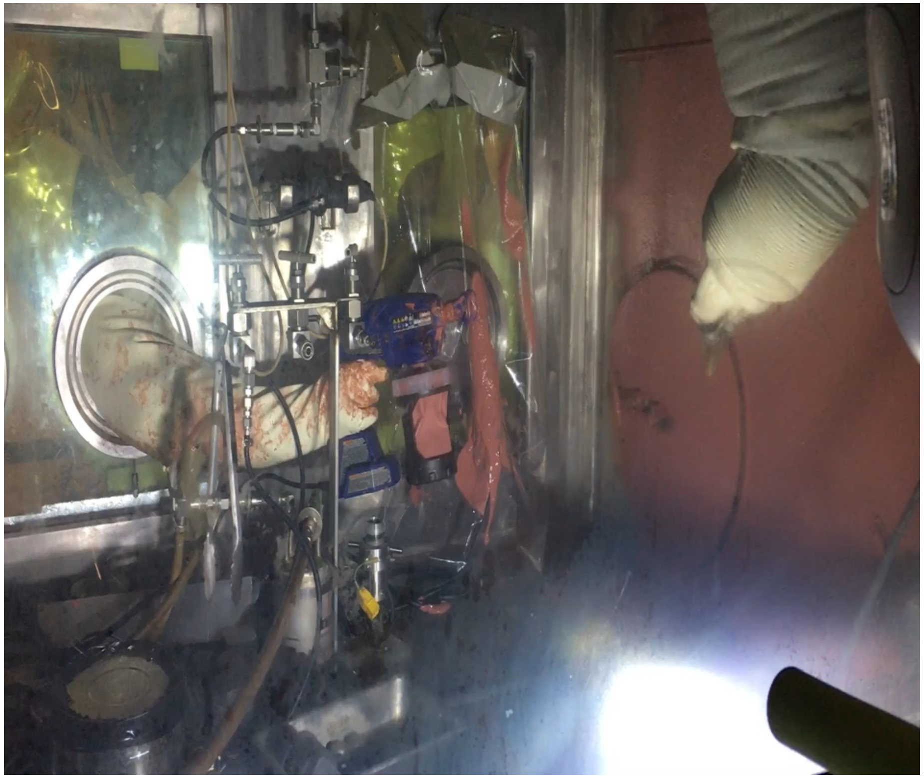
Spray Application in Entry Hood

- Spray time for entire bottle with one person swapping between gloves ~4 minutes
  - Spray approx. half the bottle in one position
  - Monitor out of gloves
  - Spray rest of bottle in second position
  - Monitor out of gloves
  - Change cartridge in first position and repeat
- Difficulty in establishing a "midline" for the spray led to overspray into area by position 2

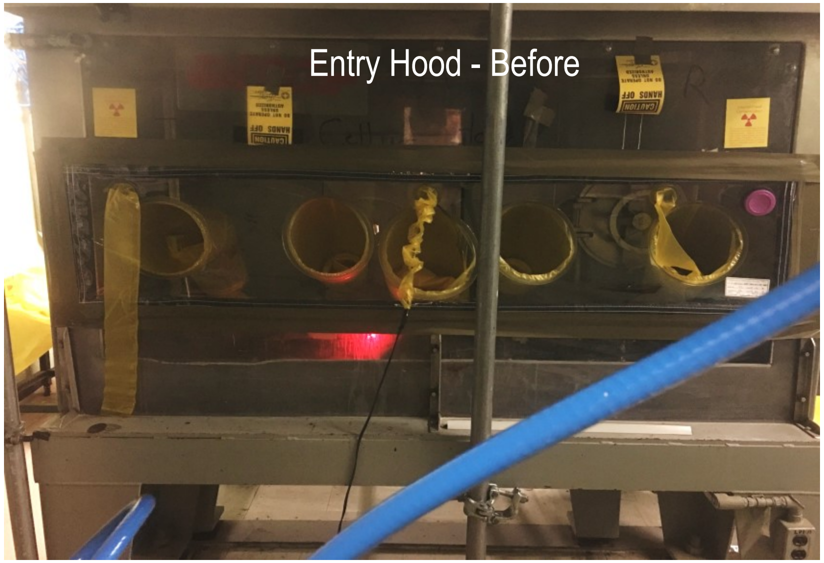
- Target: 3.2 mm
- Positions inside grey box are on the plastic masking for square door
- Overspray form first position showed for points in black box
- Total placed on wall was 1.5 gal over two spray sessions
- Sprayed 1 gallon more on targeted areas

[redacted]

Spray Application of FD – Mockup Vs. Cell 7



Before and After – Spray Application



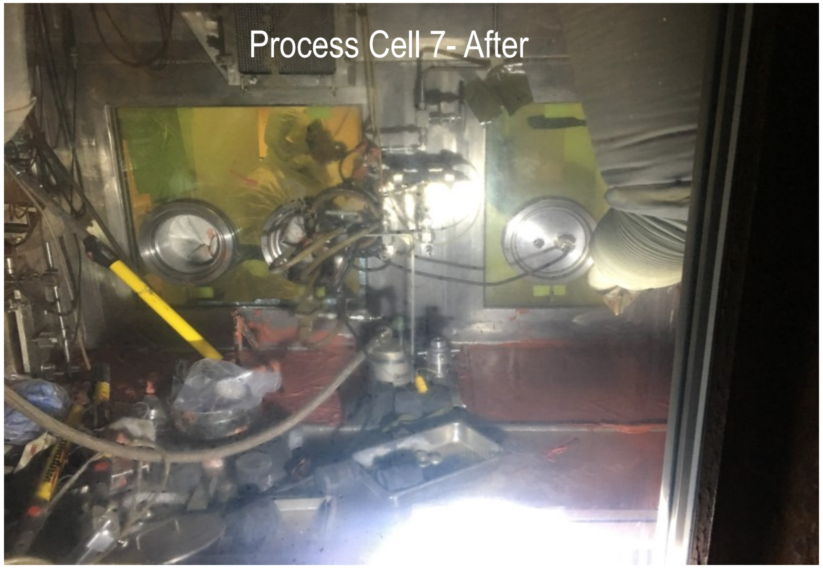
Spread Application in Entry Hood

- Spread time for 4 bottles (~1 gal/ 3.78 L) with one person pouring and spreading entire area ~10 minutes
  - Pour all bottles across floor
  - Monitor out of gloves
  - Introduce spreading tool into entry hood
  - Spread along entirety until visually level
- Difficulty around protrusions (electrical outlet housing, various protrusions from back wall)
  - Could be solved with smaller spreading tool

- Target: <u>3.2 mm</u>
- All measured points were over the requisite thickness after only one application
- Total placed on floor was 1 gallon over one spread period

Spread Application of FD – Cell 7

Before and After –Spread Application

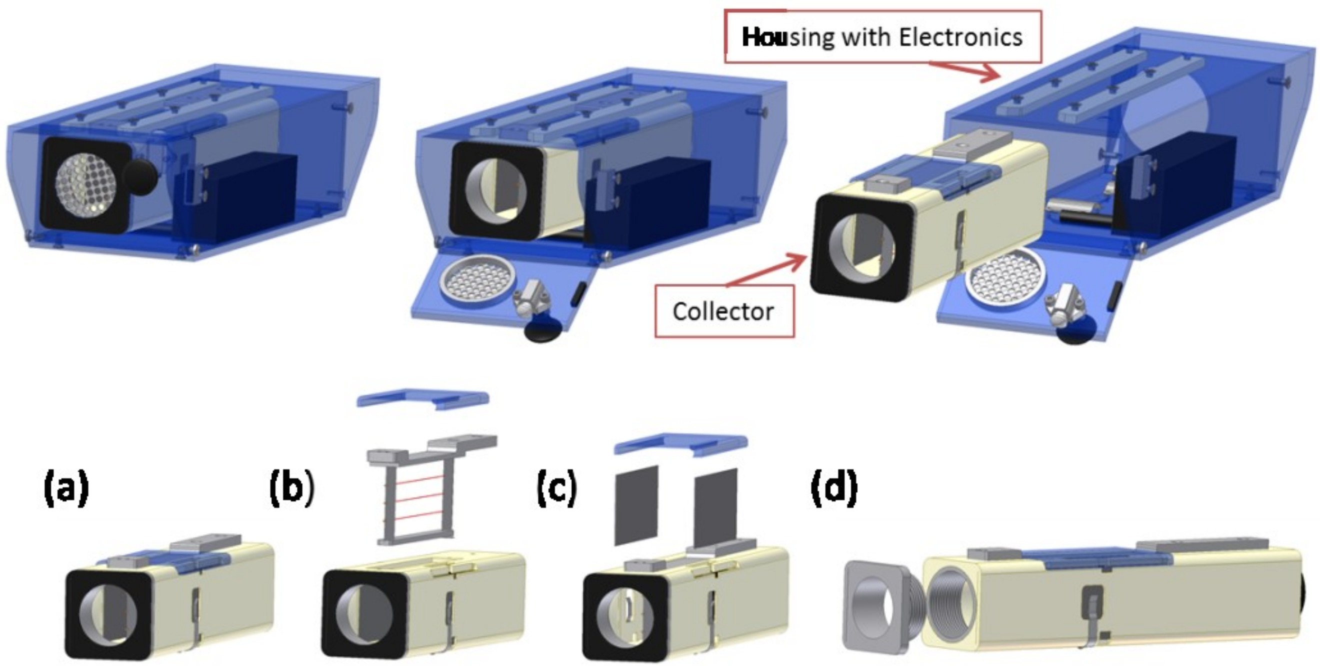


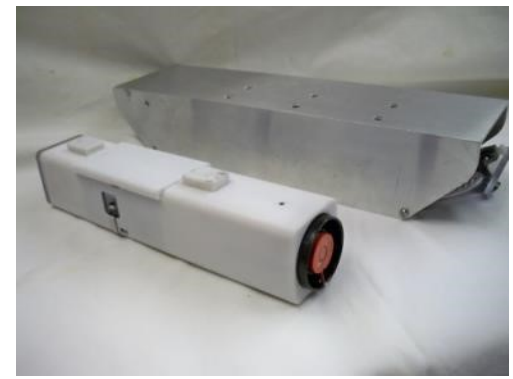
Skill Sets Required to Complete Project

- Initiative – at the start, no defined standards or metrics for fixative materials
- Technical Skills
  - Identifying different testing metrics
  - Testing different application methods
  - Navigate Safety Basis requirements for nuclear facilities
  - Comply with Waste Acceptance Criteria for all waste generated inside nuclear facility

- Coordination
  - FIU
  - Technicians
  - Safety personnel
  - Management
  - DOE HQ
  - Multiple site coordination
- Technical writing
  - Test plans
  - Safety documentation
  - Interim and final reporting documentation

Aerosol Contaminant Extractor (ACE) 2.0





Features

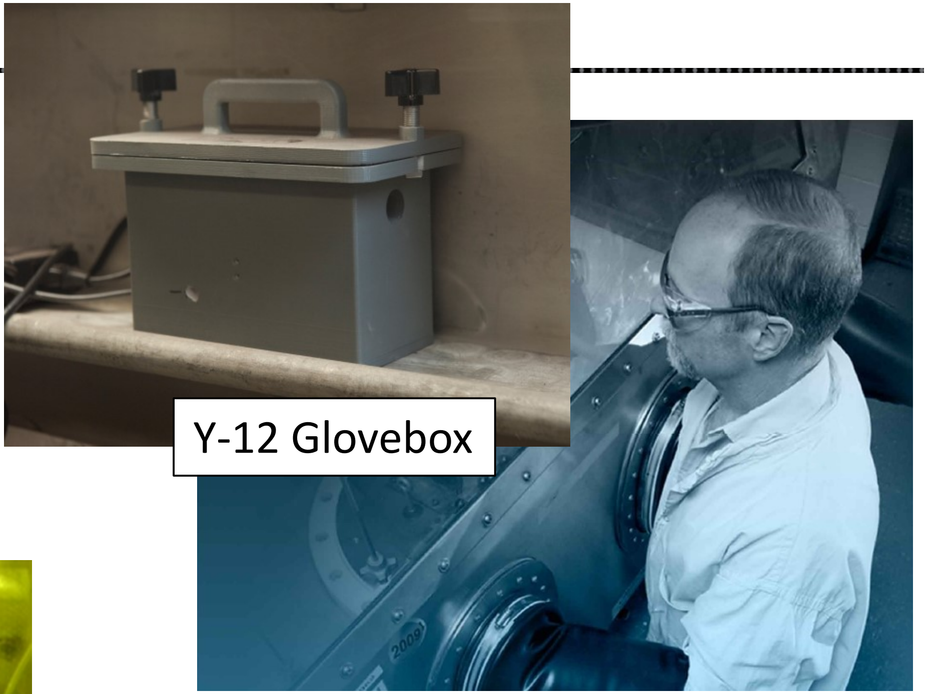
- Single Stage - Variable Voltage (up to 18kV) Electrostatic Collector
- Variable Fan Speed – up to 490 LPM
- Interchangeable Inlet Nozzle
- Remote Start/Controls
- Data Logging Capabilities
- Modular Components
- Easy substrate removal. Particles are collected on two conductive substrates

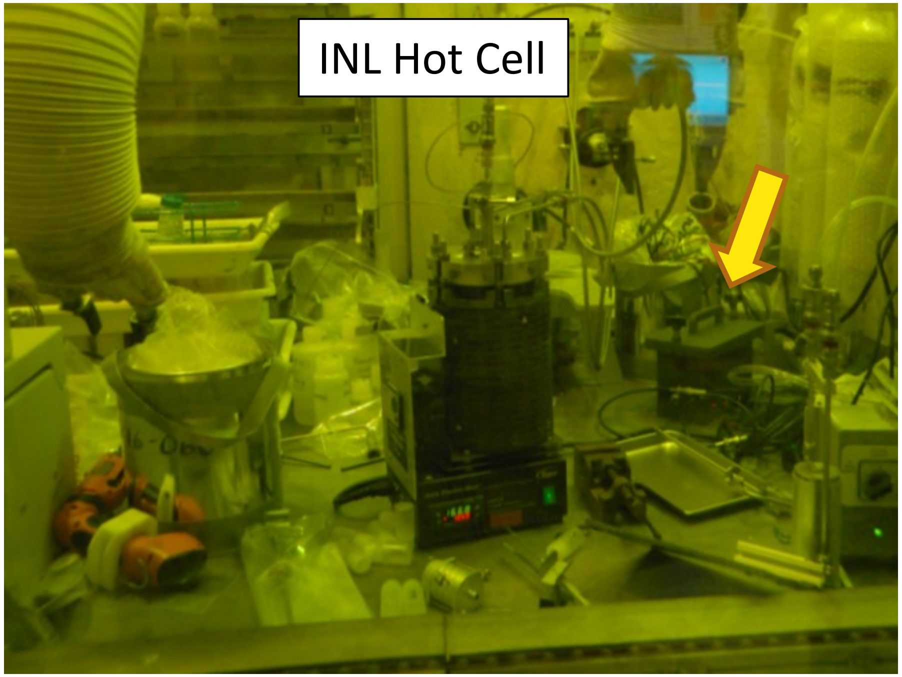
% Collection Efficiency (15 kV)
(Flow Rate – 300 LPM)

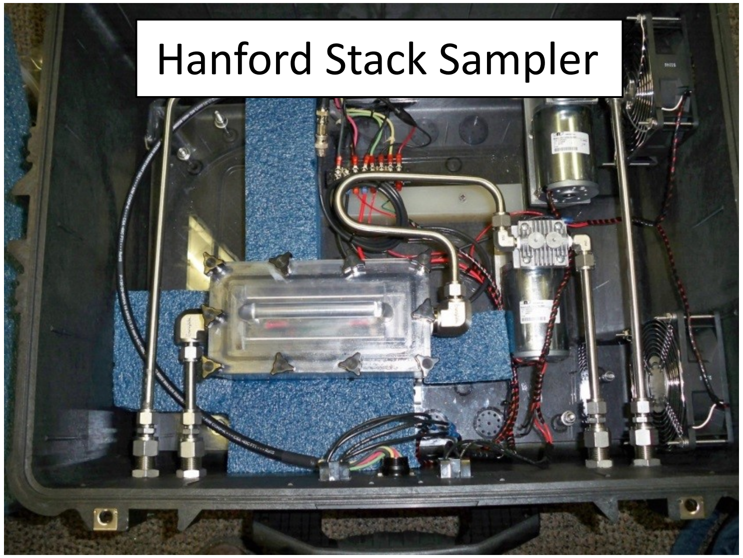
Size Range (nm)	ACE	ACE 2.0
1000-2000	12.8	38.4
500-1000	10.3	33.1
200-500	8.6	26.3

MiniACE

- Designed for Hot Cell use
- 28.3 L/min flow rate (1 ft^3/min)
- Successfully employed at multiple DOE sites



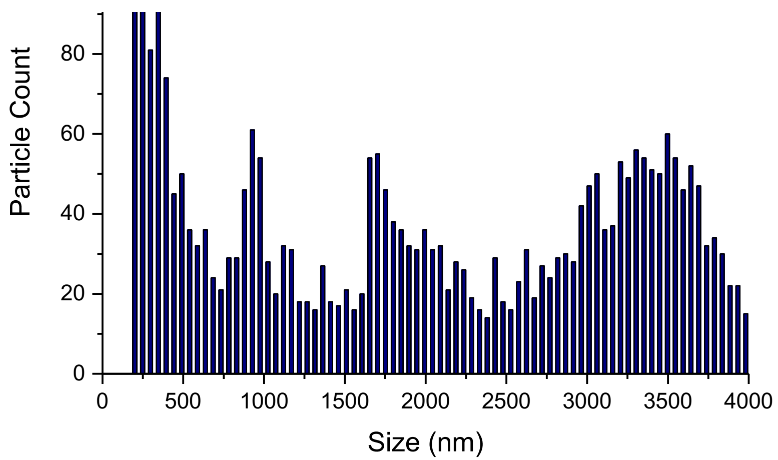




SRNL's Environmental Aerosol Chamber

- Experiments performed in 1.375 cubic meter, humidity controlled chamber
- NaCl particles were generated by ultrasonic nebulization of aqueous solutions of NaCl
    - Water removed via. heating and desiccation
- Airborne NaCl concentrations were measured in real-time for particle sizes ranging from 0.2 to 4.0 µm using a LasX-II particle counter (TSI, Inc.)

Plot of airborne NaCl Particle Size vs. Count

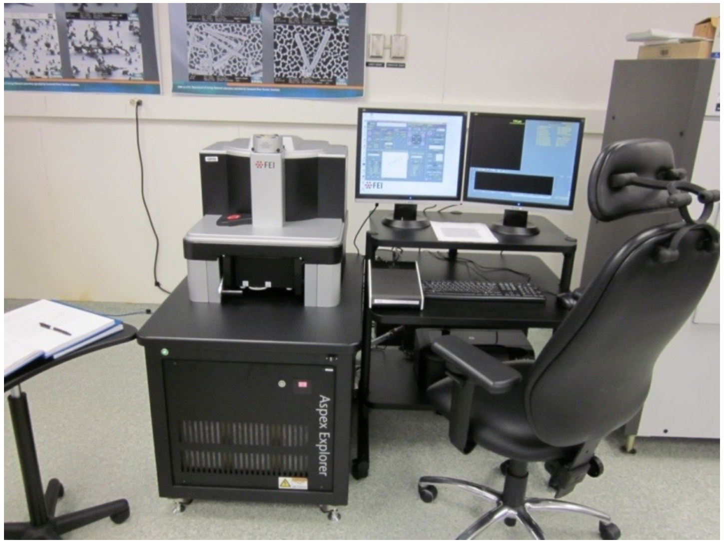


Test Chamber

Electrostatic Particle Collector Characterization

Particle Size/Distribution measurements using Computer Controlled Scanning Electron Microscopy (CCSEM)

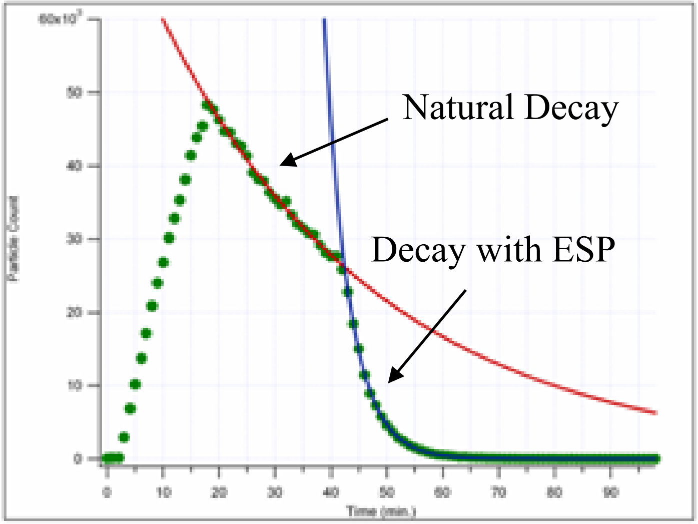
- **FEI – Aspex Explorer**
- **Aspex Perception Software (GSR)**
- **Smallest detectable feature size:**
  - 200 nm (mag. 900x)
  - Working Distance: ~15 mm



Collection efficiency assessment based on Particle Depletion Dynamics (PDD)

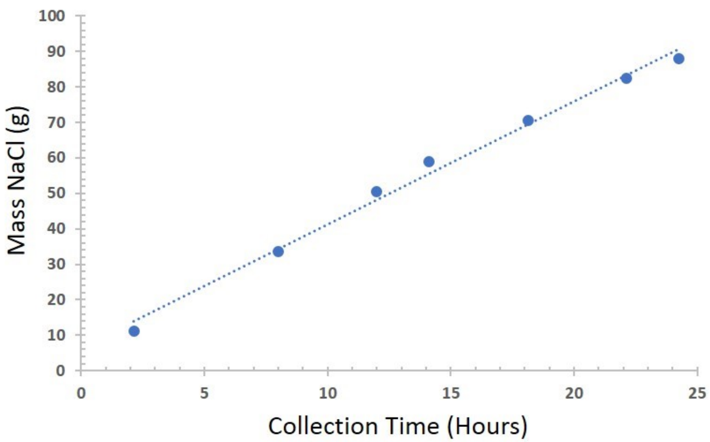
- **Monitor change in aerosol decay rate in enclosed system**
- **Collection efficiencies are determined by comparing decay rates with and without an activated EPC**

Plot of aerosol decay rate

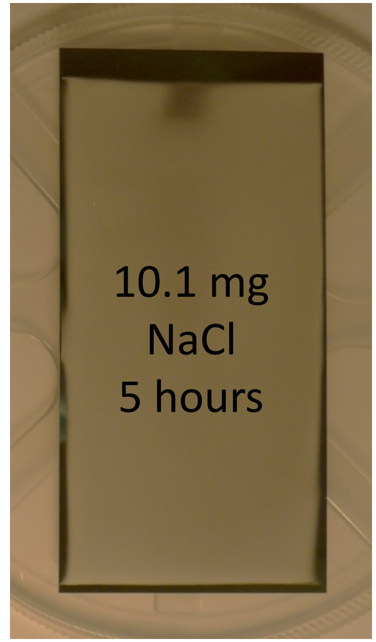


EPC Particle Loading Experiments with Aerosolized NaCl

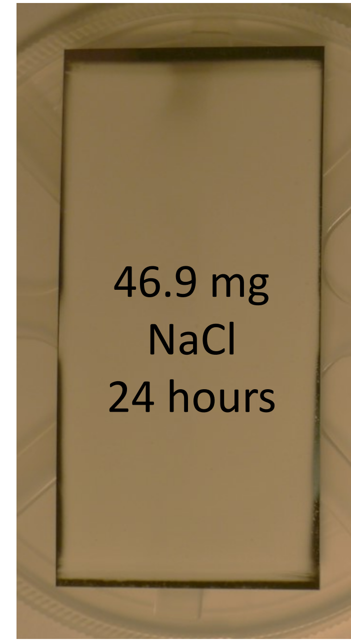
- NaCl aerosol continuously injected into aerosol chamber with active EPC
- NaCl loading determined by mass balance
- Collection rate showed little change after 24 hours
  - 2 mg NaCl per hour per substrate
- SEM images reveal bare substrate still exposed after 24 hour collection period

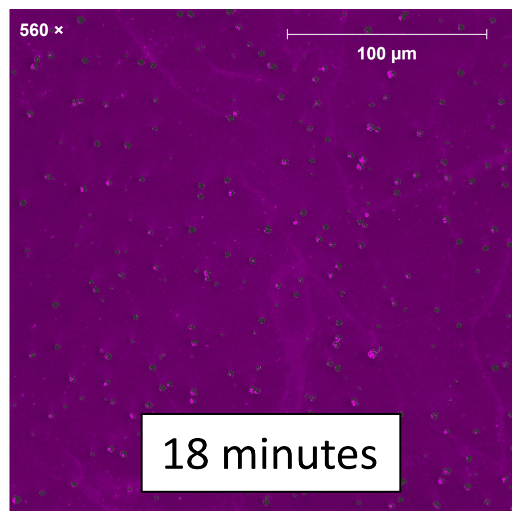


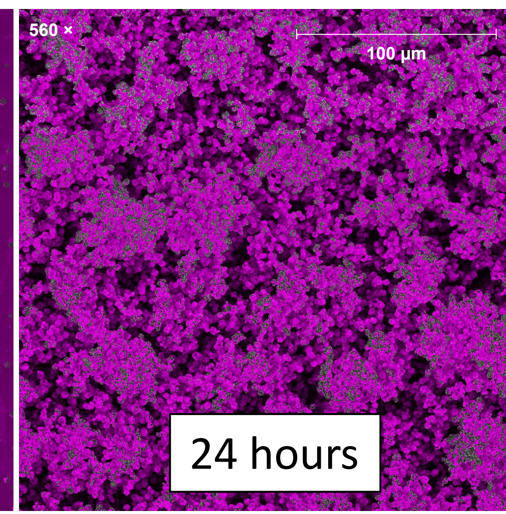
NaCl particles collected per time



Direction of Airflow







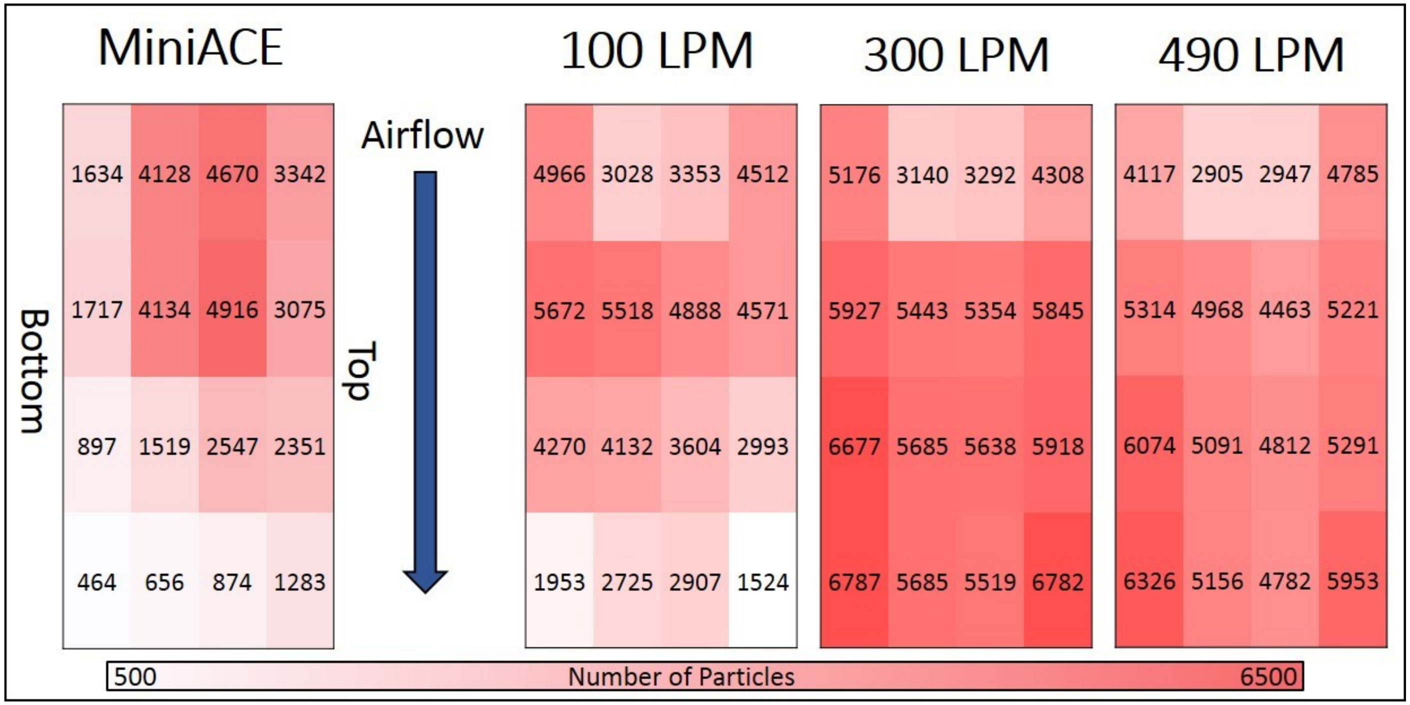
EPC Collection Efficiency Results

	Flow Rate (L/min)	Collector Efficiency (%)
ACE 2.0	100	54
ACE 2.0	300	29
ACE 2.0	490	13
MiniACE	28	> 90%

- **Collection efficiencies decrease with increasing flow rates**
- **Number of particles collected per unit time generally increases with increased flow rate even with lower collection efficiencies**
- **MiniACE static flow rate of 28.3 L/min was too low for efficiency determination using PDD**
  – Collection efficiency was inferred using CCSEM measurements

Spatial Distribution of Collected Particulates

NaCl particles collected per mm^2 divided into 4x4 grids for different EPCs and flow rates



- **Particle density maps reveal the spatial distribution of NaCl particles across the entire 1.75 x 3.55” collection substrates**
- **Particles collect near inlet with lower flow rates**

Spatial Distribution of Collected Particulates: Size vs. EPC Flowrate

200–500 nm

100 L/min				300 L/min				490 L/min			
135	104	98	136	120	62	69	131	105	66	77	134
153	165	158	172	131	133	116	136	140	138	106	117
182	164	124	142	135	131	106	147	120	110	105	132
194	217	159	144	293	173	144	237	279	160	170	213

60 Number of Particles 250

0.5 –1.0 µm

100 L/min				300 L/min				490 L/min			
459	339	363	480	395	269	256	411	337	254	254	453
502	583	505	529	431	528	474	482	426	487	426	476
576	505	488	431	497	436	446	522	489	411	347	472
413	524	480	366	940	644	559	813	789	564	521	742

250 Number of Particles 900

1.0–2.0 µm

100 L/min				300 L/min				490 L/min			
1040	628	677	980	1046	617	660	948	859	561	559	1038
1198	1222	1091	1014	1201	1085	1123	1199	1029	968	956	1030
1058	1004	919	820	1359	1142	1141	1310	1251	1043	1000	1164
556	752	714	477	1609	1291	1193	1485	1383	1071	1014	1329

500 Number of Particles 1600

> 2.0 µm

100 L/min				300 L/min				490 L/min			
3332	1957	2215	2916	3616	2193	2308	2819	2815	2025	2058	3160
3819	3547	3135	2856	4164	3698	3641	4027	3719	3376	2975	3598
2455	2460	2074	1599	4687	3977	3945	3939	4215	3527	3361	3524
791	1233	1553	537	3946	3577	3623	4247	3875	3362	3077	3669

550 Number of Particles 4600

Proof of Concept: Airborne Pu-238 Collections at PuFF

- Two locations were selected to demonstrate the use of an ACE to collect Pu-238 at the Plutonium Fuel Form (PuFF) Facility:
  - Process Cell 7
    - Prior to interior of cell being disturbed
    - After fixative has been applied to cell walls
  - Inside entry hood
    - Prior to interior of cell being disturbed
    - After fixative has been applied to cell walls

Process Cell 7

Entry Hood

Proof of Concept: Airborne Pu-238 Collections at PuFF

- ACE ran undisturbed for 24 hours for each collection
- 300 L/min flow rate
    - Total volume of air sampled: 432,000 Liters
- Alpha measurements of collection substrates performed by a Bicron
    - 1000 dpm alpha measured for both collection sites prior to and after fixative application
    - Cell 7 was declared to have <1 gram Pu-238 prior to work start
    - Entry Hood was not cleaned prior to work start and declared to have ~3 grams Pu-238
- Similar uptake results for both a decontaminated cell and a contaminated entry hood show that surface contamination is not the only concern
    - Pu-238 is known to self suspend in air due to alpha recoil
    - As work is performed in these areas, material is suspended in air
    - Electrostatic precipitation offers a method for collecting this suspended contamination and plating onto a solid substrate for ease of disposal post job

Skill Sets Required to Complete Project

- Initiative – reaching out to responsible parties and selling idea to HQ
- Technical Skills
  - Laboratory scale testing of valid parameters
  - Understanding of facility specific parameters
    - Humidity
    - Stagnate negative pressure environment
    - Limited stirring of dust in old facility (likely very little airborne)

- Coordination
  - SRNL NS and ES Directorates
  - Technicians
  - Safety personnel
  - Management
  - DOE HQ
  - Multiple site coordination
- Technical writing
  - Test plans
  - Safety documentation
  - Interim and final Documentation