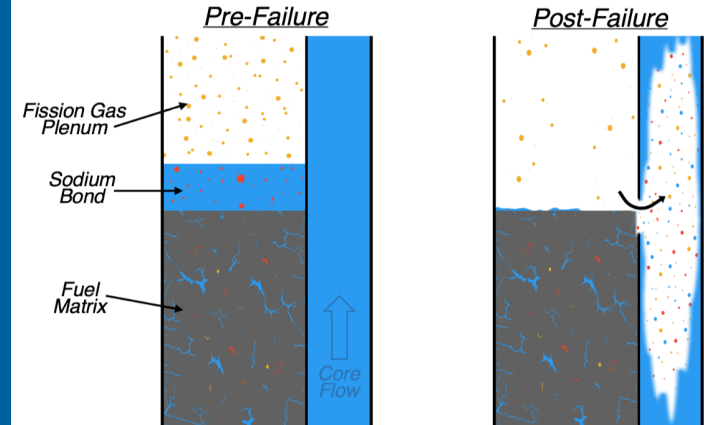


MECHANISTIC SOURCE TERM



Dave Grabaskas
Argonne National Laboratory

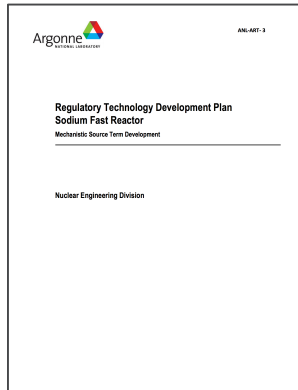
March 27, 2019
Fast Reactor Technology Training
U.S. Nuclear Regulatory Commission

OBJECTIVES

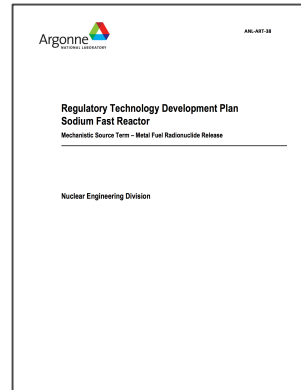
- Understand sources of radioactivity
- Understand barriers to the release of radioactivity to the environment
- Understand radionuclide transport and retention phenomena
- Understand available data (experiments, accidents, analyses)
- Understand modeling tools

MECHANISTIC SOURCE TERM ANALYSIS

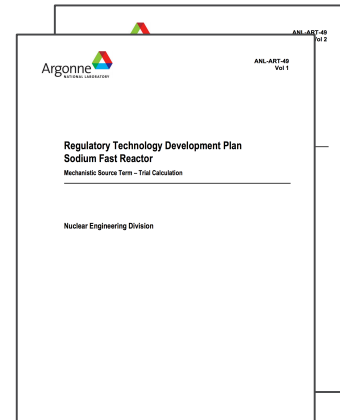
- Focus of this presentation is on metal fuel source term phenomena
 - Preferential choice of U.S. SFR vendors
 - Many phenomena after fuel failure are very similar for either metal or oxide fuel
- Source term analysis has been a recent focus of Argonne research
 - All reports are public



ANL-ART-3
Overview of SFR
source term phenomena



ANL-ART-38
Radionuclide release fractions
from failed metal fuel pins



ANL-ART-49
Mechanistic source term
trial calculation

Sources of Radioactivity



```
graph TD; A[Sources of Radioactivity] --> B[Barriers and Phenomena]; B --> C[Transport and Retention]; C --> D[Past Accidents]; D --> E[Modeling];
```

Barriers and Phenomena

Transport and Retention

Past Accidents

Modeling

Sources of Radioactivity

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graph TD; A[Sources of Radioactivity] --> B[Barriers and Phenomena]; B --> C[Transport and Retention]; C --> D[Past Accidents]; D --> E[Modeling];
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Barriers and Phenomena

Transport and Retention

Past Accidents

Modeling

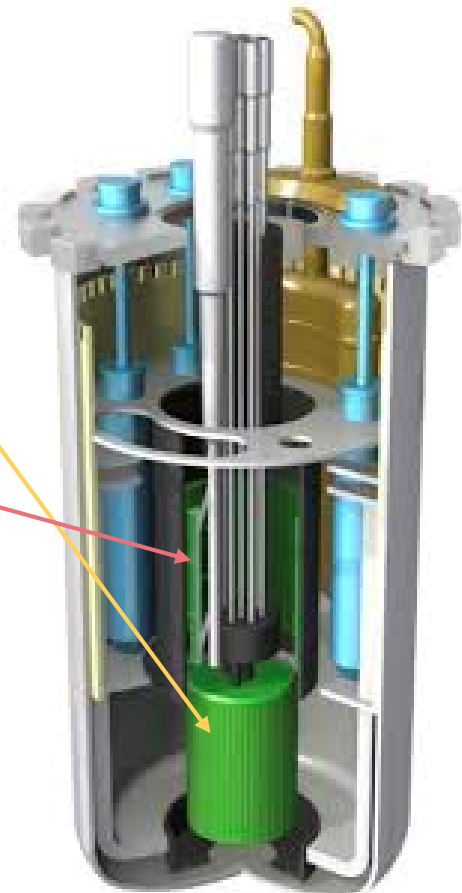
SOURCES OF RADIOACTIVITY: CORE

■ Active fuel in core

- Exact composition and burnup can vary greatly (as described in earlier lectures)
- Certain fuel failure phenomena are composition/burnup dependent

■ Other fuel in vessel

- Most pool-type SFRs provide in-vessel storage
- Allows preheating of fresh fuel
- Spent fuel interim storage
- Exact location in vessel varies by design



SOURCES OF RADIOACTIVITY: PRIMARY Na

■ Activated Sodium

- Natural sodium is entirely Na-23
- Na-24 (half-life ~15 hours) and Na-22 (half-life ~2.6 yrs) are produced during irradiation
- Shielding is used to minimize intermediate sodium activation in IHXs

■ Corrosion Products

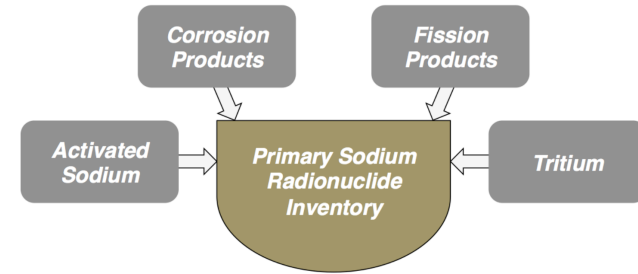
- Creation and activation of **Co-58**, **Co-60**, **Mn-54**, Fe-59, Ta-128

■ Fission Products

- From failed fuel (discussed in later in presentation)

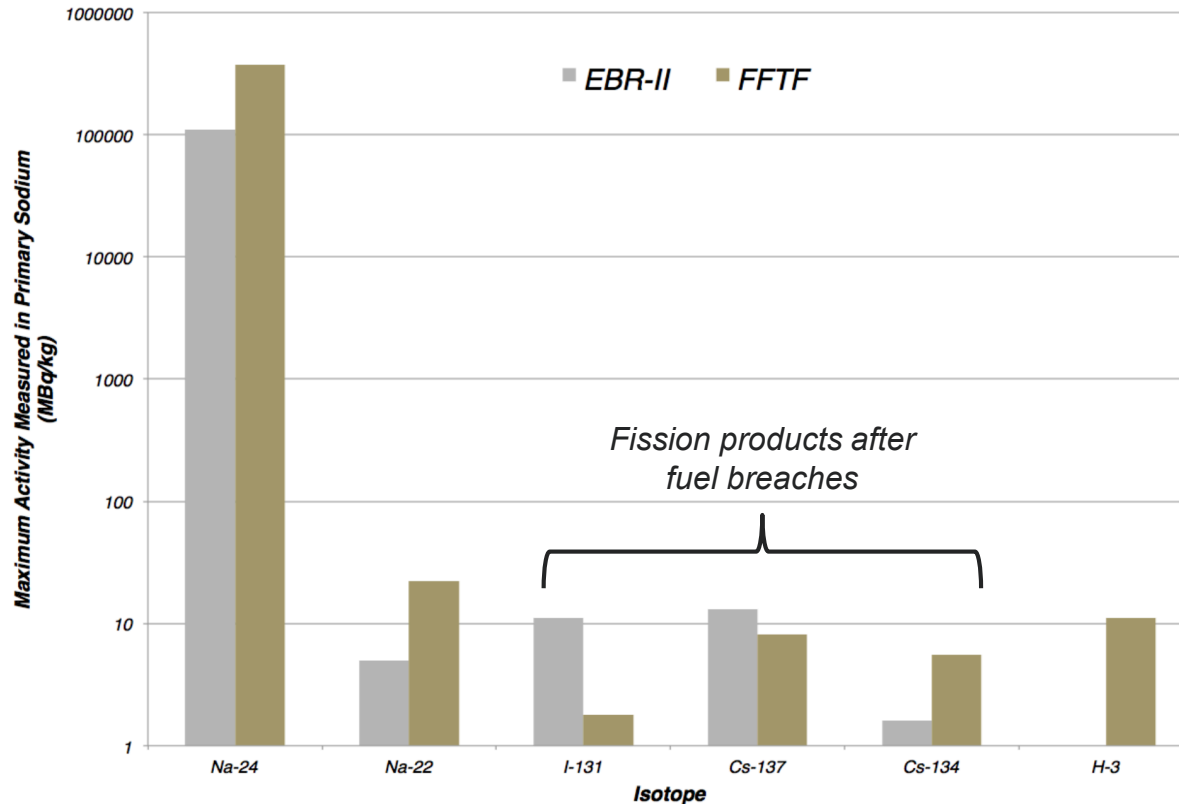
■ Tritium

- From ternary fission and boron neutron capture
- Diffuses through cladding, IHXs, steam generators, etc.



SOURCES OF RADIOACTIVITY: PRIMARY Na

Maximum activity levels recorded in primary sodium of EBR-II and FFTF



SOURCES OF RADIOACTIVITY: COVER GAS

■ Activated Sodium Decay products

- Ne-23 (half-life ~38 secs)

■ Activated Cover Gas

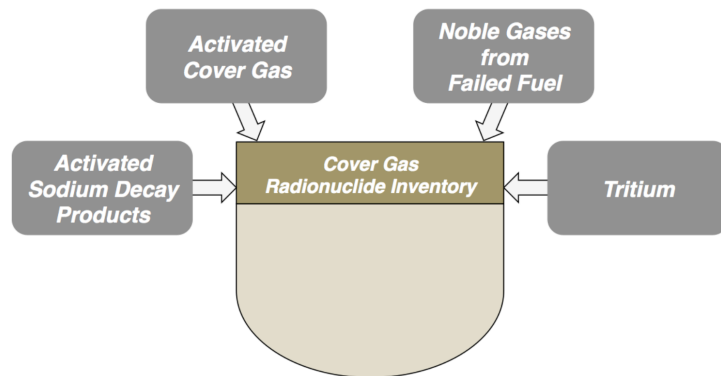
- Typically Ar-40 is used for cover gas
- Several reactions produce Ar-41 (or K-41)

■ Noble Gases (Fission Products)

- From failed fuel (discussed in later in presentation)

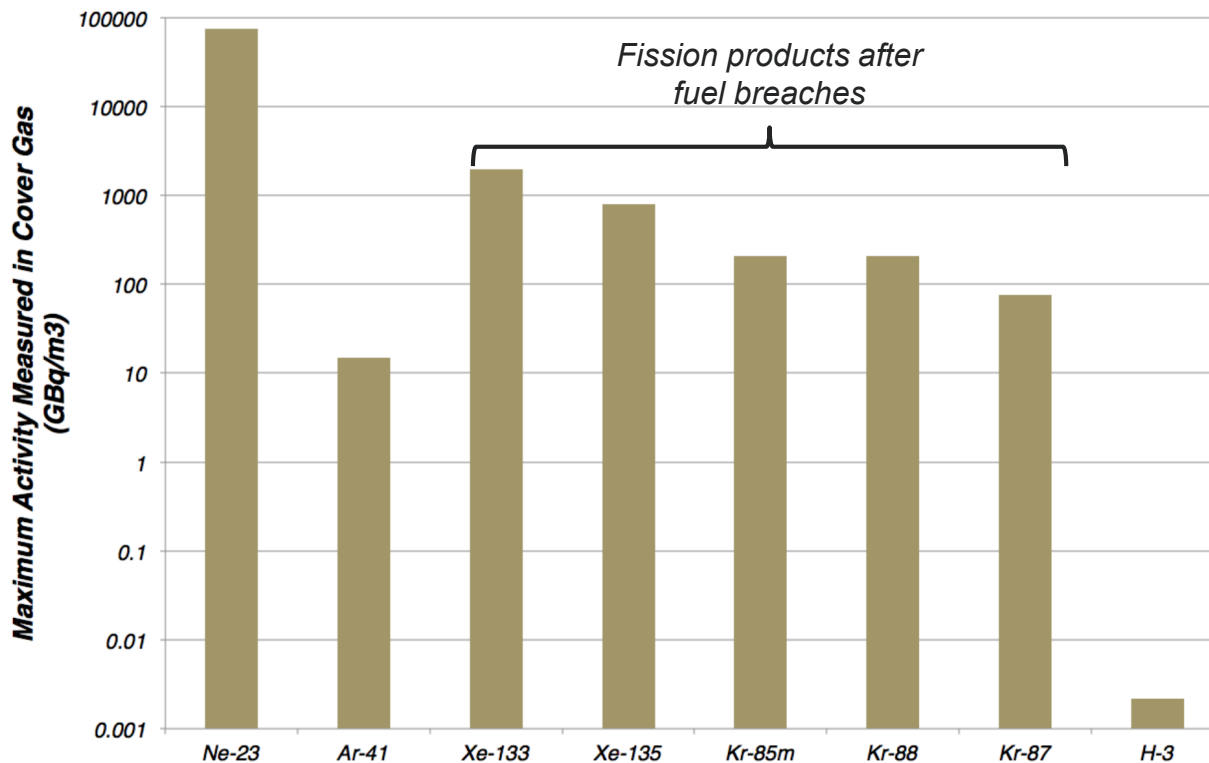
■ Tritium

- From ternary fission and boron neutron capture
- Diffuses through cladding, IHXs, steam generators, etc.



SOURCES OF RADIOACTIVITY: COVER GAS

Maximum activity levels recorded in cover gas of FFTF



SOURCES OF RADIOACTIVITY: EX-VESSEL

■ Spent Fuel Movement

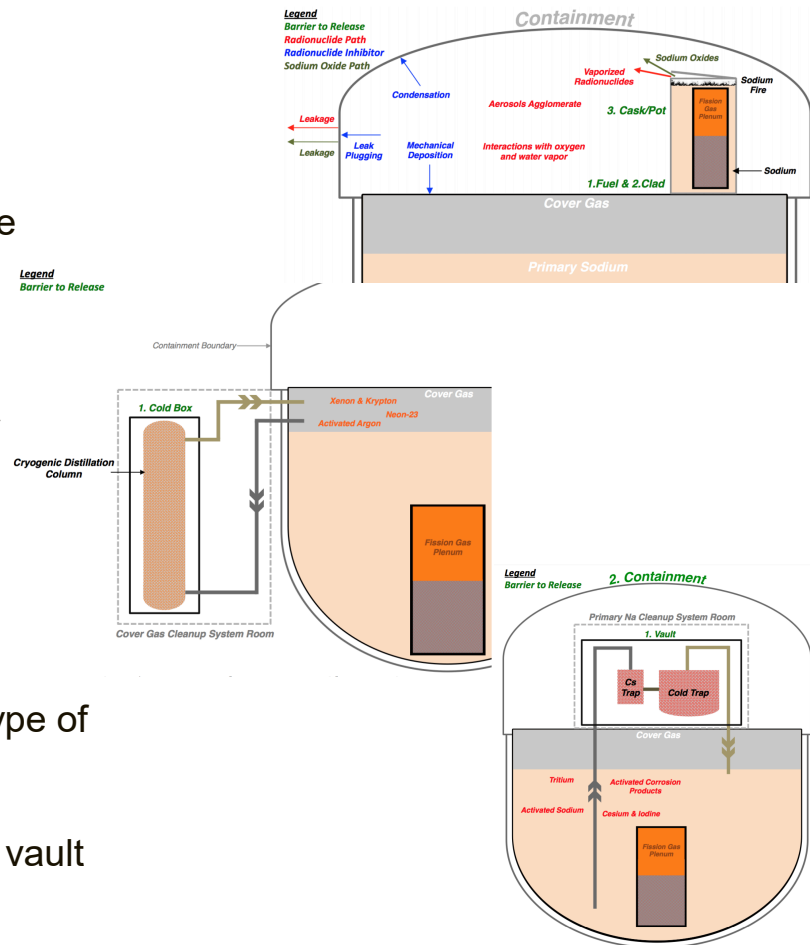
- Fuel may be moved in sodium or inert gas casks
- A loss of cooling could result in fuel failure
- A breach of a sodium-bearing cask could result in a fire

■ Cover Gas Cleanup System

- Designs vary but past reactors have used cryogenic distillation to condense noble gases and allow time for decay
- A loss of cooling or system break may result in re-volatilization of the noble gases

■ Primary Sodium Cleanup System

- Designs vary but most contain a cold trap and some type of cesium trap
- A leak and sodium fire is likely the most challenging scenario for the system but it is typically enclosed in a vault or sealed room to mitigate such accidents



Sources of Radioactivity

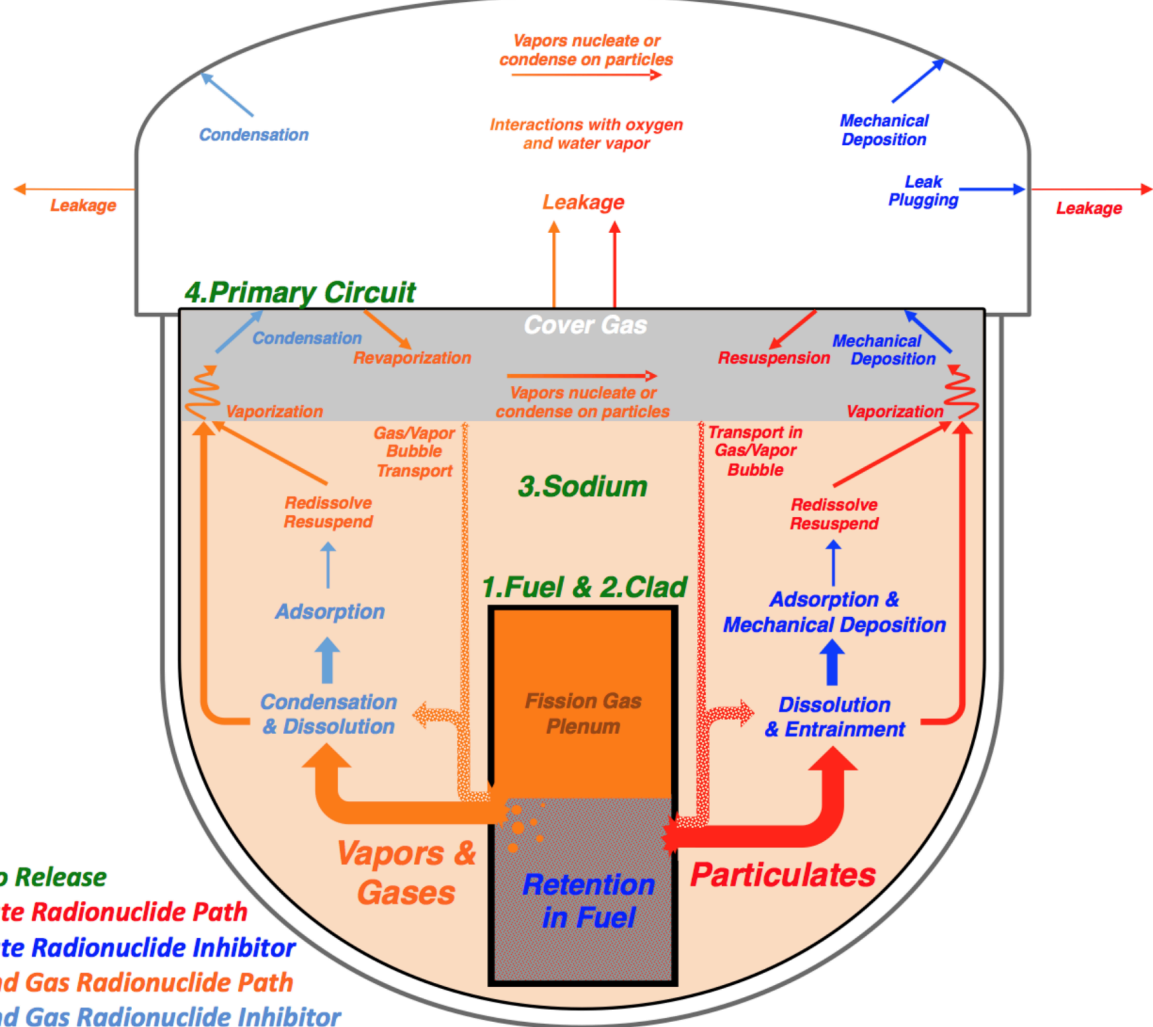
Barriers and Phenomena

Transport and Retention

Past Accidents

Modeling

5. Containment



Legend

Barrier to Release

Particulate Radionuclide Path

Particulate Radionuclide Inhibitor

Vapor and Gas Radionuclide Path

Vapor and Gas Radionuclide Inhibitor

KEY DIFFERENCES: LWRs(OXIDE) FUEL

■ Release Pathways

- For LWRs, during primary system breaks the water level drop in the core region can create a direct transport pathway from failed fuel to containment
- For SFRs, the primary sodium pool provides an additional, significant retention mechanism as the loss of sodium below the core level is extremely unlikely

■ Containment Loading

- For LWRs, containment pressures during accident sequences can be high due to the failure of high pressure piping and potential hydrogen explosions
- For SFRs, postulated accident scenarios do not involve the rupture of a high pressure boundary and no hydrogen production, which means significantly less pressure loading on containment

■ Behavior of Iodine

- Unlike oxide fuel, in metal fuel iodine can bond with uranium to form uranium iodides, which retain the majority of iodine in the fuel matrix during most postulated accident scenarios
- Iodine that is released from the fuel matrix is typically in the form of stable sodium iodide (NaI), not volatile molecular iodine (I_2)

Sources of Radioactivity



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graph TD; A[Sources of Radioactivity] --> B[Barriers and Phenomena]; B --> C[Transport and Retention]; C --> D[Past Accidents]; D --> E[Modeling];
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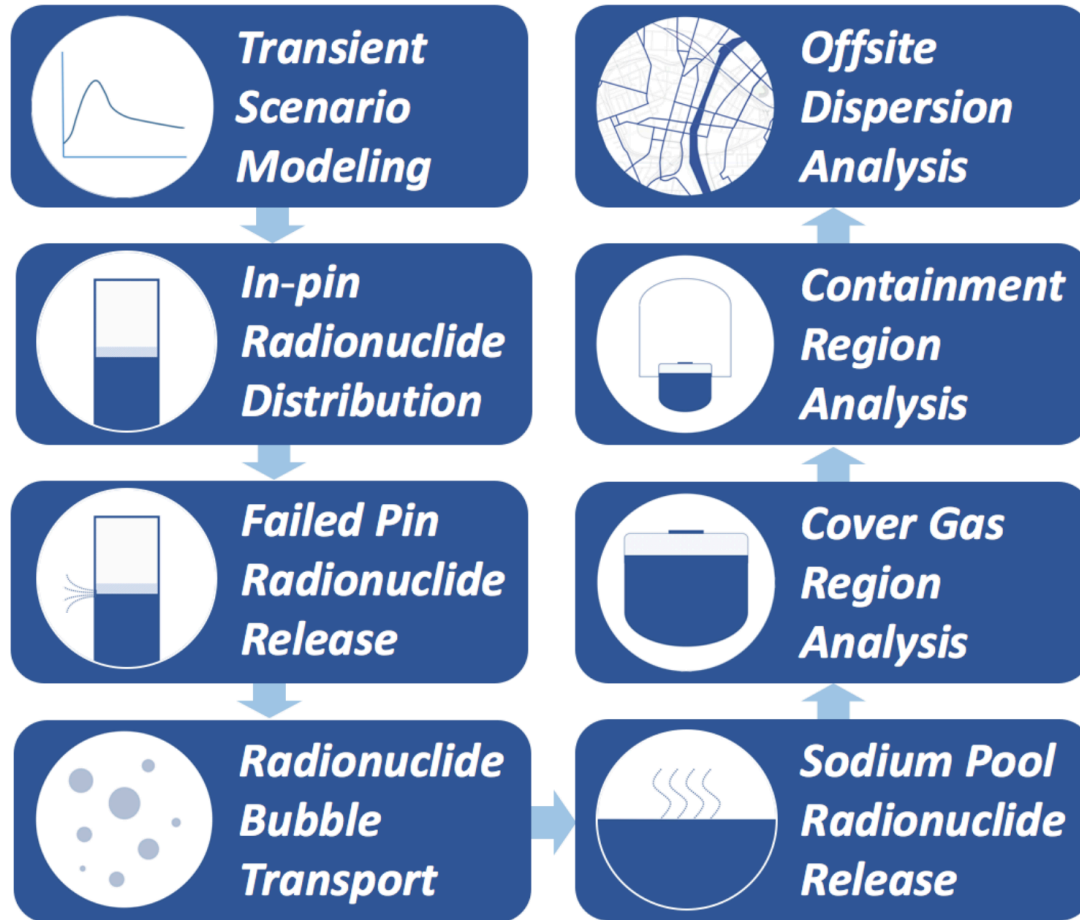
Barriers and Phenomena

Transport and Retention

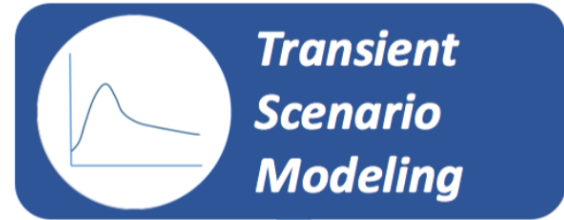
Past Accidents

Modeling

MECHANISTIC SOURCE TERM ANALYSIS



TRANSIENT ANALYSIS



■ Core Damage Event Sequences

- Two most credible, although still very unlikely, core damage scenarios
- Typically these scenarios are in the beyond design basis/residual risk frequency range

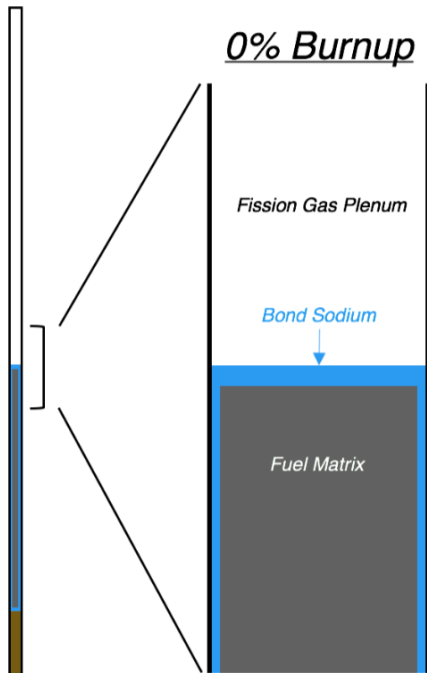
1) Long-Term Loss of Decay Heat Removal

- Reactor shutdowns successfully, but there is an extended (>24 hours) complete loss of decay heat removal
- Fuel and primary sodium temperatures slowly increase, ΔT between fuel and sodium is small
- Failure of the metal fuel pins occurs due to eutectic penetration of the cladding (no melting of the fuel matrix)

2) Rapid Power-to-Flow Mismatch

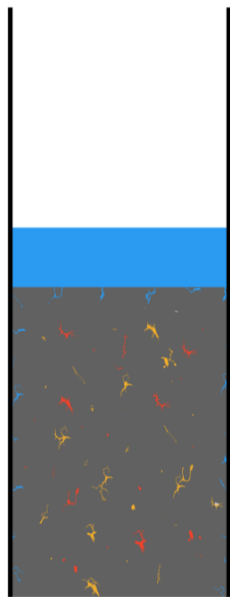
- May be caused by an extreme transient overpower, grossly reduced pump coastdown, or large flow blockage
- Fuel temperature increases rapidly, causing fuel matrix melting then cladding failure, before termination of transient within seconds to minutes of initiation
- Sodium pool temperatures remain approximately nominal

RADIONUCLIDE DISTRIBUTION



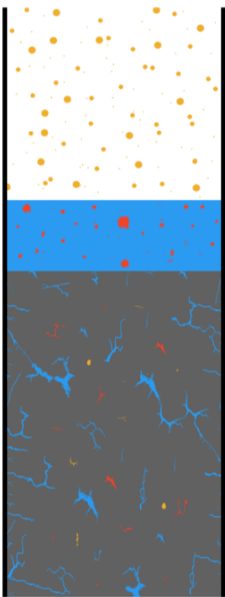
The fresh fuel matrix is surrounded by *bond sodium*.

~1% Burnup



*Gaseous radionuclides and non-gaseous radionuclides are formed, causing the fuel matrix to expand and come in contact with the cladding while extruding *bond sodium* to the plenum region.*

>2% Burnup



*The porosity of the fuel matrix interconnects, allowing *gaseous radionuclides* to transport to the plenum region, while some *non-gaseous radionuclides* migrate to the *bond sodium*. *Bond sodium* logging within the fuel matrix porosity also occurs.*

Fuel Pin

RADIONUCLIDE DISTRIBUTION



■ Radionuclide Migration

- During irradiation, the metal fuel matrix expands and become porous
- In addition, radionuclide (fission product) migration occurs within the fuel pins
- See [ANL-ART-38](#) for a description of the migration behavior of all radionuclides

■ Noble Gases

- The majority of xenon and krypton inventory migrates to the fission gas plenum once the fuel porosity interconnects, which occurs around 1-2% burnup

■ Cesium

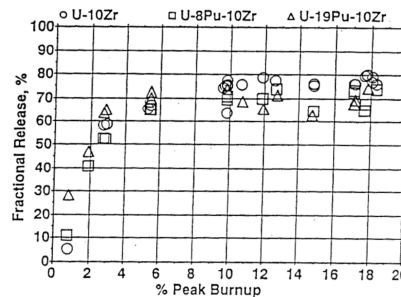
- Cesium migrates to the bond sodium in the fission gas plenum, the migrated fraction increases with burnup level

■ Iodine

- Majority of iodine remains in fuel matrix as uranium iodides, small fraction migrates to bond sodium

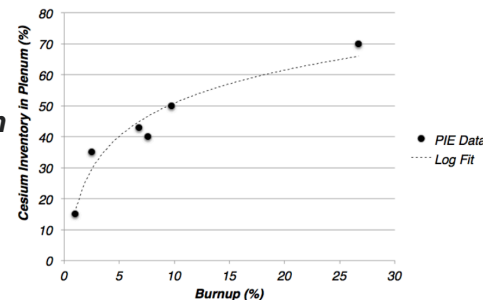
■ Strontium

- Depending on fuel type, significant migration to the bond sodium may occur



Noble Gas Migration vs. Burnup

Cesium Migration vs. Burnup



RADIONUCLIDE DISTRIBUTION

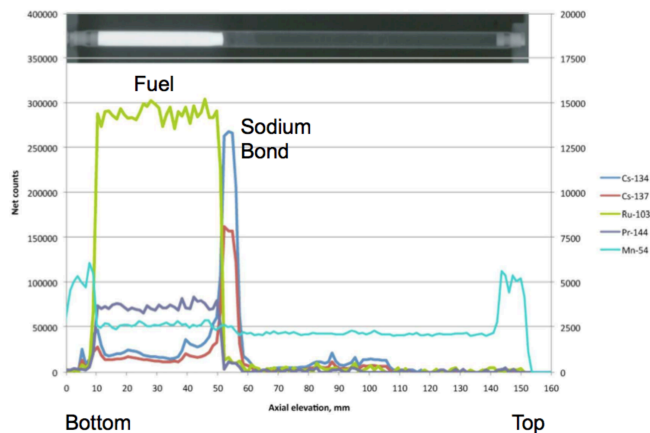


■ Significant Validation Data Available

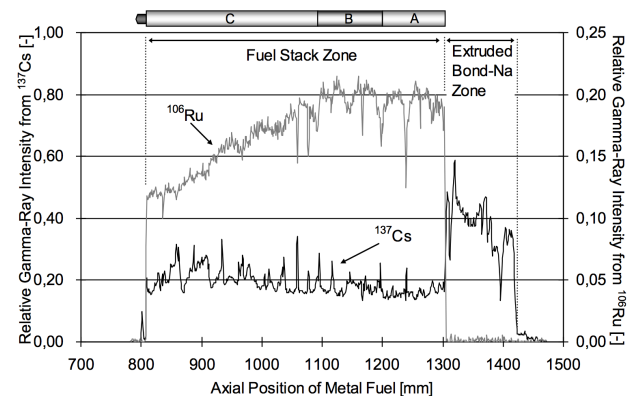
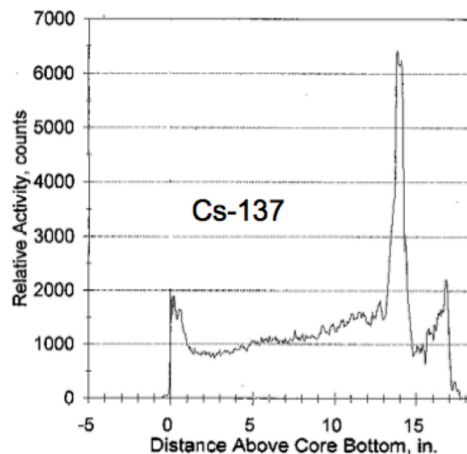
- See [ANL-ART-38](#) for complete details

■ Post-Irradiation Examination (PIE)

- Analysis tracked migration of many major fission products



Axial Gamma Scans



RADIONUCLIDE RELEASE



■ Radionuclide Release Phenomena

- With the failure of the cladding, the contents of the fission gas plenum and bond sodium are likely to be released from the fuel pin
- Depending on the temperature and fuel conditions, additional radionuclides may be released from the fuel matrix
- The following two slides examine two potential cases of fuel failure, aligning with the two types of core damage event sequences described on slide 18

RADIONUCLIDE RELEASE

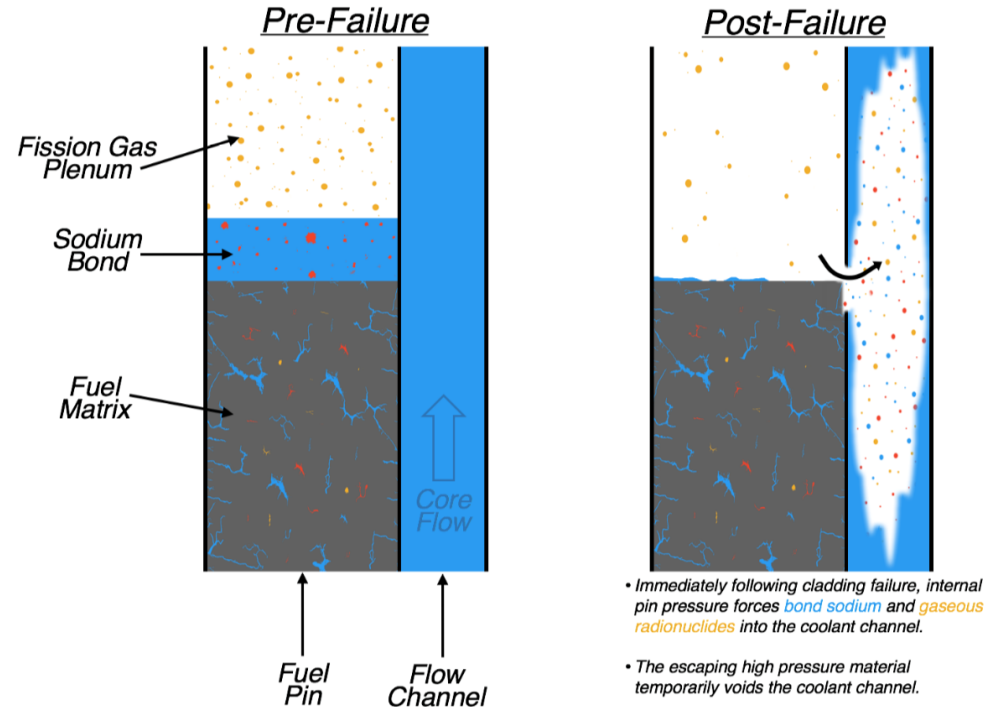


■ Failure Due to Eutectic Penetration

- Can occur during long-term elevated temperature events (loss of decay heat removal)
- No melting of the fuel matrix
- Cladding failure occurs near the top of the active fuel region since it is the hottest spot

■ Radionuclide Release

- Contents of the fission gas plenum and bond sodium are released and will likely create a temporary void in the sodium due to the high internal pressure of the fuel pin
- Additional radionuclide releases from the fuel matrix are small as temperatures are below the fuel melting temperature and radionuclide vaporization temperatures



RADIONUCLIDE RELEASE

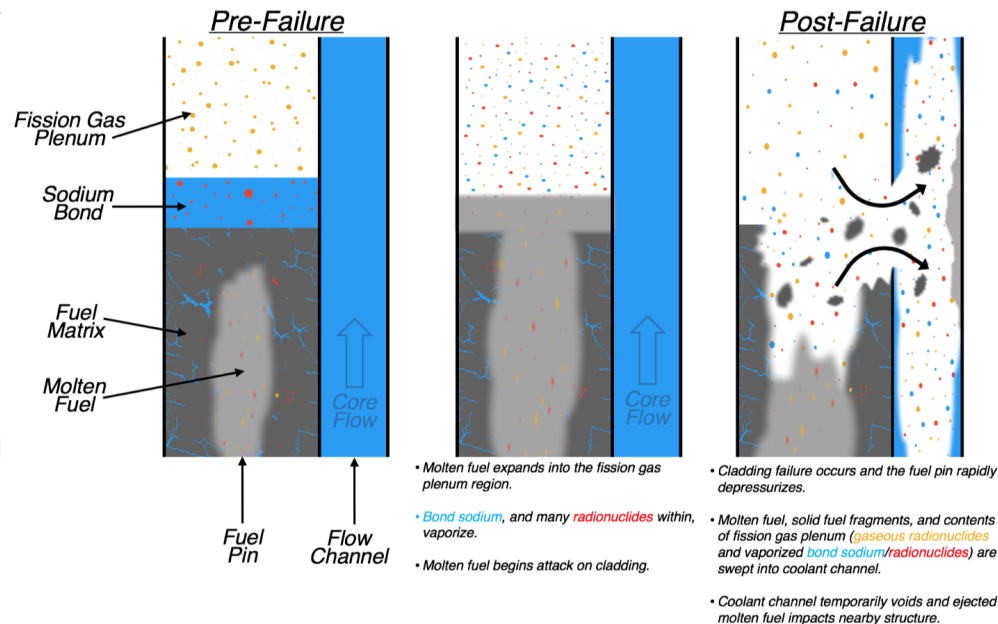


■ Failure Due to Fuel Melting

- Can occur during rapid power-to-flow mismatches
- Depending on fuel design, molten fuel may first enter the fission gas plenum before interacting with the cladding, resulting in pin failure

■ Radionuclide Release

- Contents of the fission gas plenum, bond sodium, and molten fuel are released and will create a temporary void in the sodium due to the high internal pressure of the fuel pin
- Radionuclide releases in addition to those in the fission gas plenum and bond sodium can occur due to the high temperature of the fuel matrix (exact release depends on burnup and temperature)



RADIONUCLIDE RELEASE



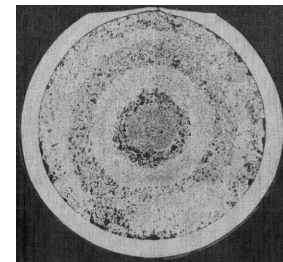
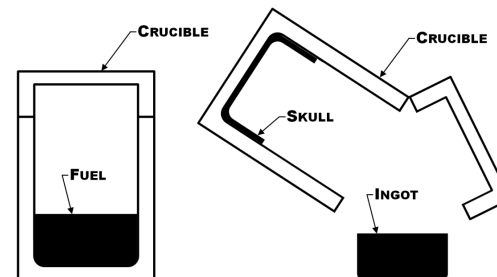
■ Significant Validation Data Available

- See [ANL-ART-38](#) for complete details
-
- EBR-II Melt-Refining Reprocessing
 - Purposely melted irradiated fuel to volatize or separate fission products for fuel reprocessing
 - Specific melting times and temperatures chosen for fission product removal

 - Run-Beyond-Cladding-Breach Tests
 - Multiple pins purposefully weakened to fail during irradiation
 - Pins remained in core for extended periods (>100 days) after failure

 - Fuel Melting Experiments
 - Fuel elements melted at different time and temperatures to explore fission product release

 - Past Accidents
 - Discussed later in presentation



RADIONUCLIDE RELEASE



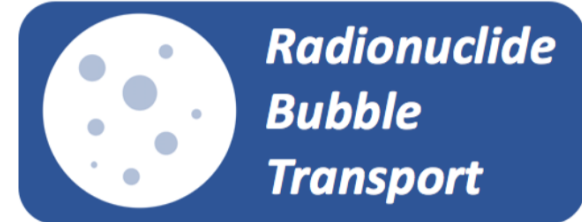
■ Example Release Fraction Table

- From ANL-ART-38

Halogens: I

Temperature	Normal Operation ~ 500°C	Eutectic Formation ~ 700°C	Fuel Melting ~ 1100°C	High Temperatures ≥ 1300°C
Release Percentage	≤ 15%	≤ 20%	≤ 30%	≤ 100%
Dependencies	Burnup	Burnup	Burnup	Time, Temp.
Uncertainty Level	Medium	Medium	Medium	Low
Sources				
Quantitative	EBR-II Reprocessing Metal Fuel PIE	SRE	Fermi 1 Melt Refining Tests Hanford Melt Tests	ORNL Melt Tests AI Melt Tests
Qualitative	EBR-II Leaker Tests		EBR-II Capsule	

BUBBLE TRANSPORT



■ Bubble Creation

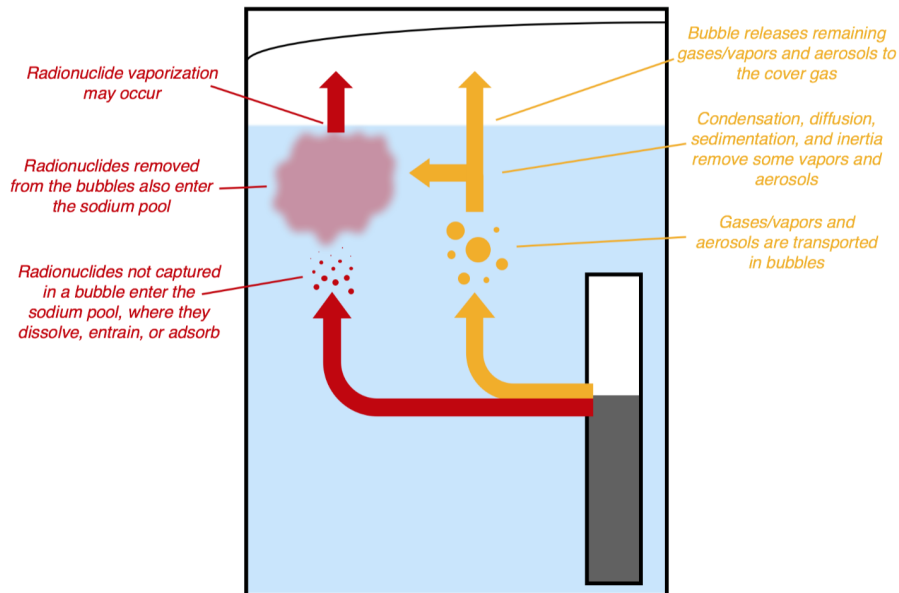
- Following fuel pin failure, noble gas bubbles are created in the primary sodium pool
- Some radionuclides become entrained in the bubbles, while others directly enter the sodium pool

■ Bubble Scrubbing

- As the bubble travels through the sodium pool, radionuclides will be removed from the bubble due to thermophoresis, diffusion, deposition, sedimentation, and condensation

■ Experimentation Ongoing

- Current university DOE-funded experiments to explore bubble transport and retention phenomena



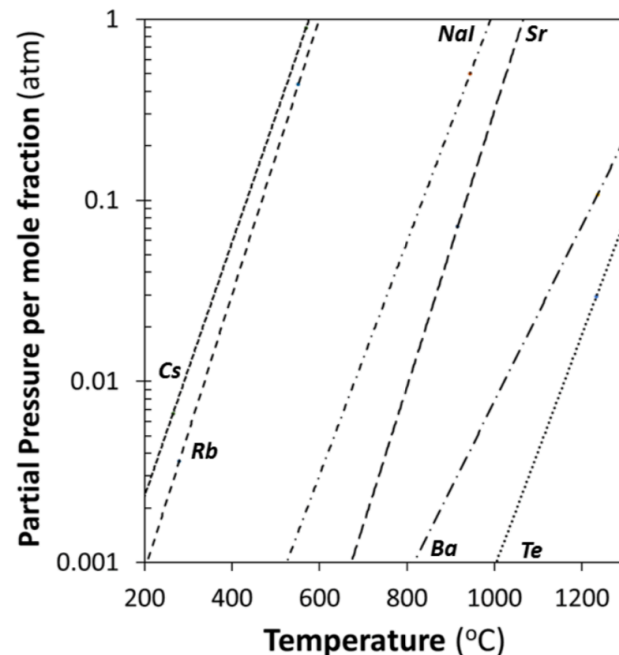
POOL VAPORIZATION

■ Vaporization

- Radionuclides that enter the sodium pool may volatilize from the pool surface and enter the cover gas region

■ Radionuclide Behavior

- Cs and Rb are the only radionuclides that are volatile at normal hot pool operating temperatures but they are very miscible with Na as they are also alkali metals
- Other radionuclides, such as NaI and Sr, require elevated pool temperatures before volatilization becomes appreciable



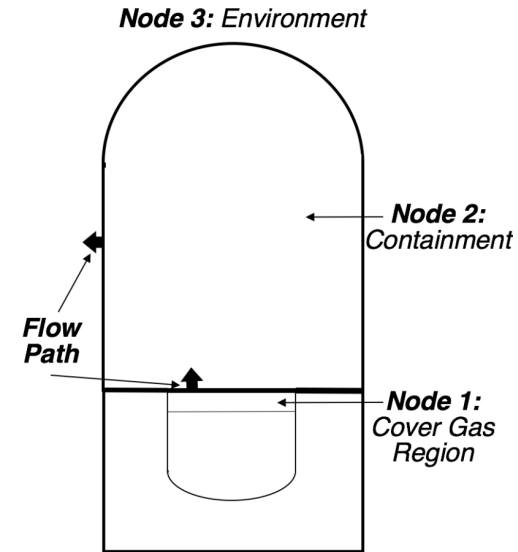
COVER GAS/CONTAINMENT

■ Cover Gas Region

- Typically an argon or helium environment
- Radionuclides in the cover gas region may be in the form of vapor/gases or aerosols
- Condensation on the cooler surfaces may occur for vapors, while aerosols may deposit on structures or the pool surface
- Radionuclides may leak out of the cover gas region through reactor head penetrations but the pressure differential is typically small
- Leakage pathways may become larger in accident scenarios with prolonged elevated temperatures of the primary system and vessel

■ Containment

- Sodium and NaI will react with the oxygen in the air
- Vapors/gases may condense in the much cooler environment
- Aerosol deposition will continue
- Leakage is expected to be small due to the low containment pressure and lack bypass mechanisms in the most credible accident scenarios



Sources of Radioactivity

```
graph TD; A[Sources of Radioactivity] --> B[Barriers and Phenomena]; B --> C[Transport and Retention]; C --> D[Past Accidents]; D --> E[Modeling];
```

Barriers and Phenomena

Transport and Retention

Past Accidents

Modeling

SODIUM REACTOR EXPERIMENT (SRE)

■ Reactor

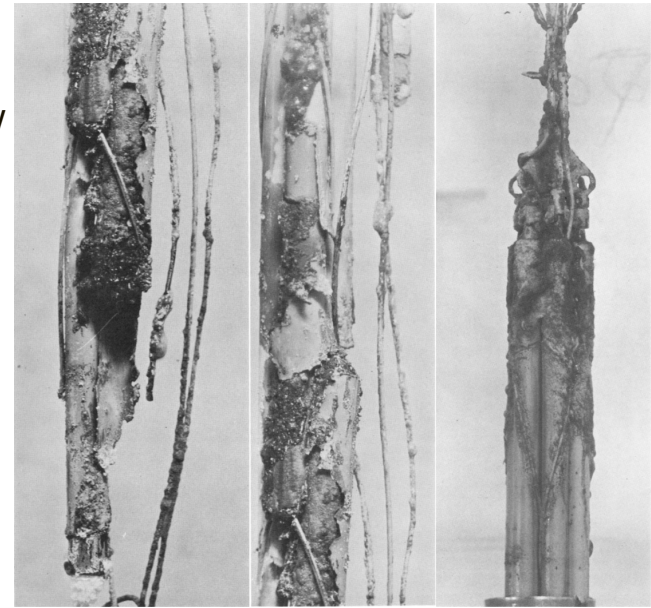
- 20MWth, graphite-moderated, sodium-cooled, thermal reactor
- Built 1957 at Santa Susana Field Laboratory in CA
- NaK-bonded, unalloyed metal fuel with SS cladding (also many experimental fuel assemblies)

■ Accident

- In 1959, a leak of primary pump lubricant (tetralin) into primary sodium occurred
- The organic compound interacted with the sodium causing a flow blockage, but the reactor continued to operate for ~10 days
- Failure of 13 of 43 fuel elements occurred due to eutectic formation, no bulk fuel melting

■ Result

- Release of fission products (Cs/I/Sr/Ba) to the primary coolant
- Only Xe/Kr detected in the cover gas
- Reactor restarted in 1960 following sodium purification and refueling operations, continued to operate until 1964



FERMI 1

■ Reactor

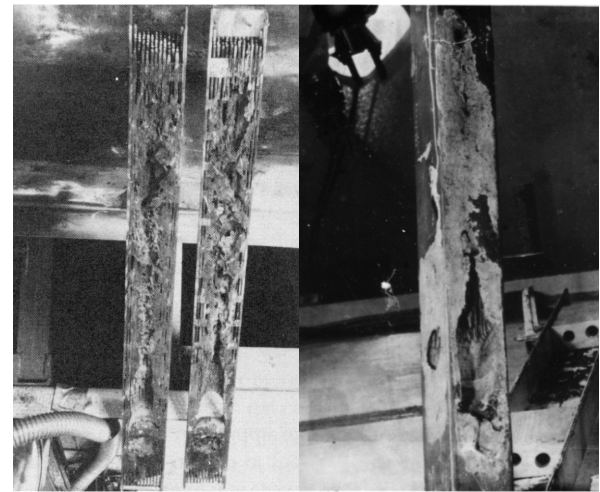
- 200MWth/66MWe, commercial sodium fast breeder reactor
- Built 1963 outside of Detroit Michigan (current site of Fermi 2)
- U-10Mo fuel with Zr cladding

■ Accident

- A late addition to the reactor design was a Zircaloy meltdown liner/spreader below the core
- In 1966, two pieces of the liner broke loose causing a flow blockage
- Significant fuel melting (~200 pins total) occurred in two assemblies where flow was <3% nominal
- Fuel damage occurred in two additional assemblies

■ Result

- Release of fission products (Cs/I/Sr/Ba) to the primary coolant
- Only Xe/Kr detected in the cover gas, minor release to containment
- Reactor restarted in 1970 and continued to operate until 1972



Sources of Radioactivity



Barriers and Phenomena

Transport and Retention

Past Accidents

Modeling

MODELING

■ A Single SFR Source Term Analysis Code Does Not Currently Exist

- There are many codes that can do parts of the calculation, such as:
 - Transient modeling and fuel damage: SAS4A/SASSYS-1
 - Bubble transport: IFR Bubble Code (Argonne internal)
 - Pool vaporization: HSC Chemistry (and similar thermodynamic equilibrium codes)
 - Cover gas and containment: MELCOR (and other aerosol codes)

■ Trial Mechanistic Source Term Analysis (ANL-ART-49)

- Demonstrated that a mechanistic analysis could be conducted with current tools
- Some models, such as in-pin radionuclide migration and radionuclide release from failed fuel pins, are primarily data-driven
- Uncertainties exist but were managed through uncertainty/sensitivity analyses
- High priority uncertainties are currently being addressed, such as bubble transport experiments

QUESTIONS?