# Mechanistic Source Term Determination

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*Changing the World's Energy Future*

**iNL**
**Idaho National Laboratory**

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# Mechanistic Source Term Determination

CNSC Seminar

Idaho National Laboratory

# Course Objectives

- Understand the function and performance of the different barriers to radionuclide release in HTGRs, how these are incorporated into reactor design and safety analyses, and approaches to estimate radionuclide release under specific reactor conditions

The Training Course delivered to the NRC in 2010 included a module discussing radionuclide behavior in HTGRs (Module 13). You are encouraged to review that course material for additional details.

# Outline

- Barriers to radionuclide (RN) release in high-temperature gas-cooled reactors (HTGRs)

- Radionuclide Design criteria

- Computational tools to predict radionuclide release and existing data

- Approach to estimate source term from HTGRs

# Radionuclide Barriers

- HTGR designs employ multiple radionuclide release barriers
  - Fuel kernels
  - Particle coatings (most important barrier)
  - Fuel-element matrix and fuel-element graphite (prismatic reactor)
  - Primary coolant pressure boundary
  - Reactor building (RB)
- These multiple radionuclide barriers provide defense in depth

Fuel Kernel (UCO, UO$_2$)
Porous Carbon Buffer
Inner Pyrolytic Carbon (IPyC)
Silicon Carbide
Outer Pyrolytic Carbon (OPyC)
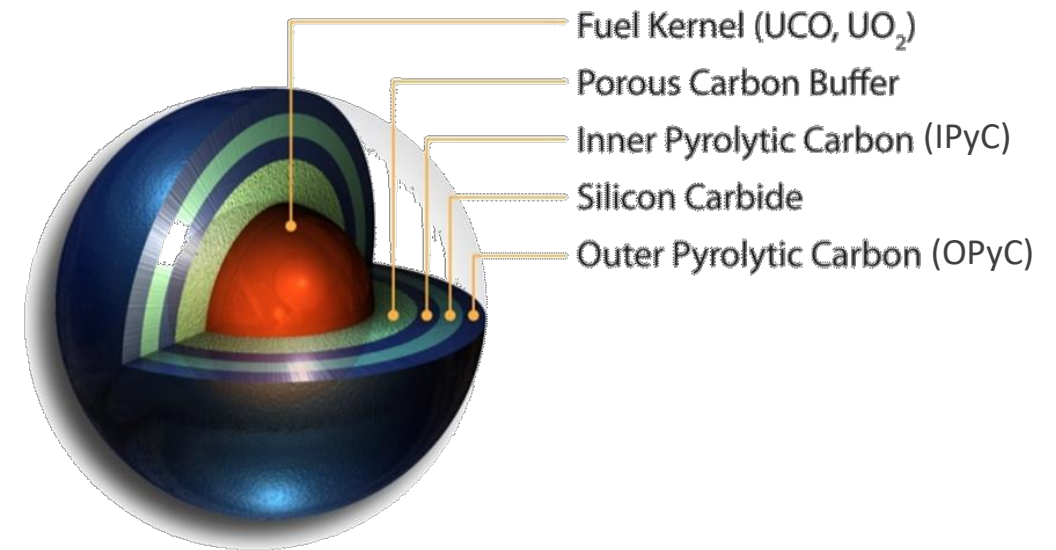
# HTGR Radionuclide Sources and Pathways
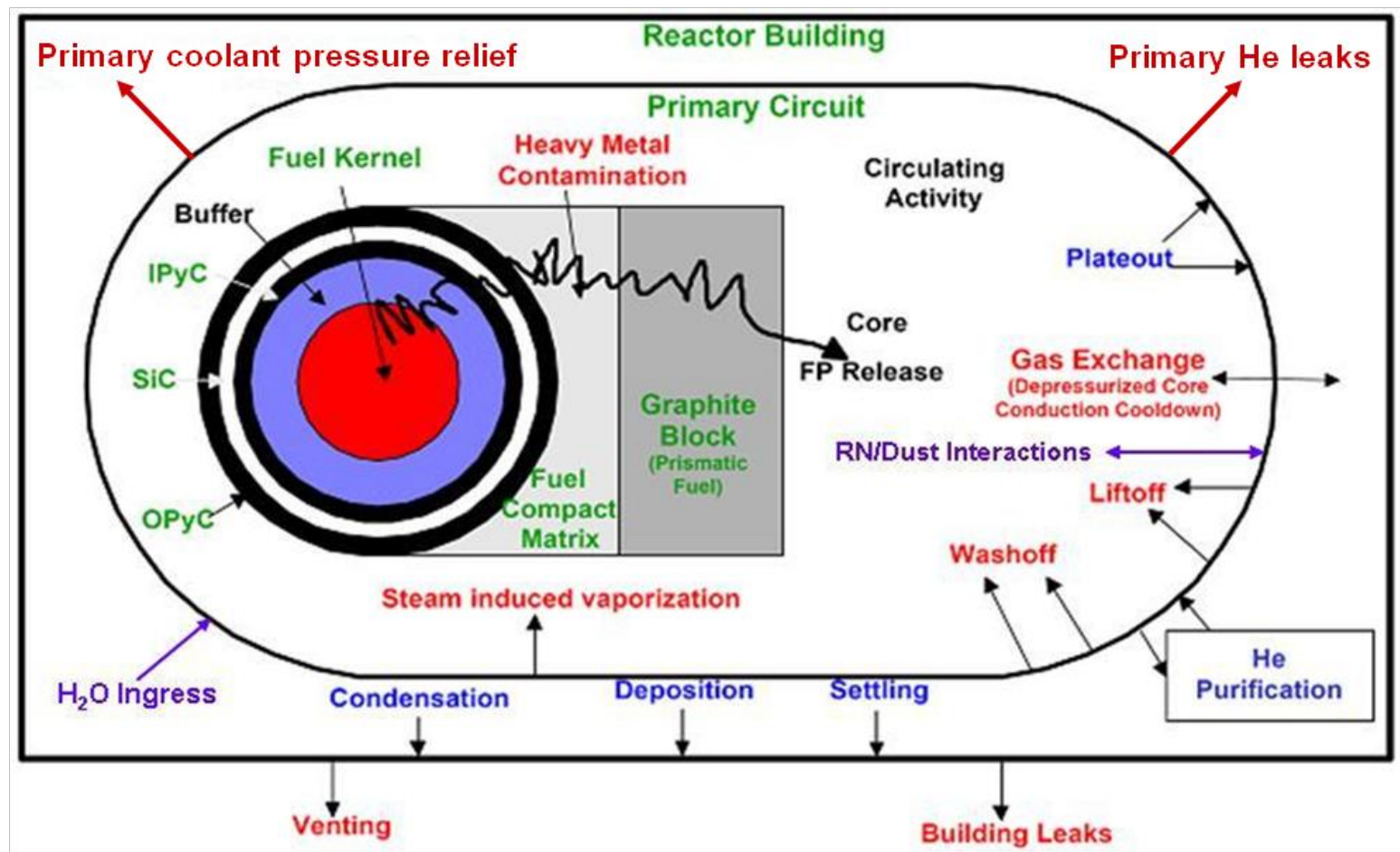
# Radionuclide Release Barrier: Fuel Kernel

- Potential release mechanisms
  - Fission recoil
  - Diffusion
  - Hydrolysis (reaction with $H_2O$)

- Controlling parameters
  - Fuel temperatures
  - Time
  - $H_2O$ concentration
  - Burnup

- Barrier performance
  - Fractional gas release function of time/temperature history
  - Increased gas release in case of hydrolysis
  - Partial diffusive release of volatile fission metals (Ag, Cs > Eu, Sr)
  - Other radionuclides, including actinides, very well retained
  - $UO_2$ expected to stabilize certain elements to a greater extent than UCO (Sr, Eu)



**Buffer**

**Kernel**

AGR-1 UCO fuel kernel, 11.3% FIMA
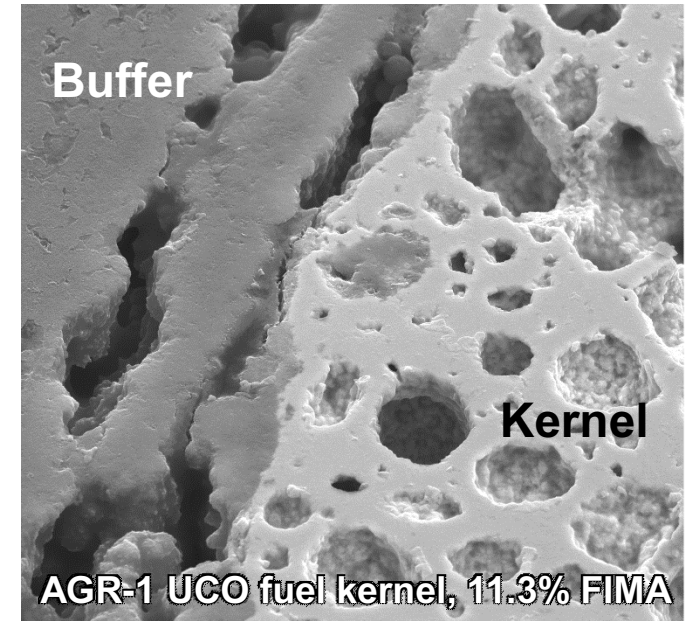
# Fission Gas Release

- Short-lived fission gas release in-pile evaluated using the "release-rate-to-birth-rate" (R/B) ratio

- Release is a function of element (Kr, Xe), isotope half life and fuel temperature

$$R_p \approx \frac{3}{x} = 3 \left[ \frac{D_0 e^{-\frac{E_a}{RT}}}{\lambda a^2} \right]^n$$

- Gas release calculated using a Booth model

- Various experiments have been performed to determine the R/B ratio for exposed kernels (including recent AGR-3/4 irradiation)

- Isotopes commonly measured:

Kr-85m
Kr-87
Kr-88
Kr-89
Kr-90

Xe-131m
Xe-133
Xe-135
Xe-135m
Xe-137
Xe-138
Xe-139
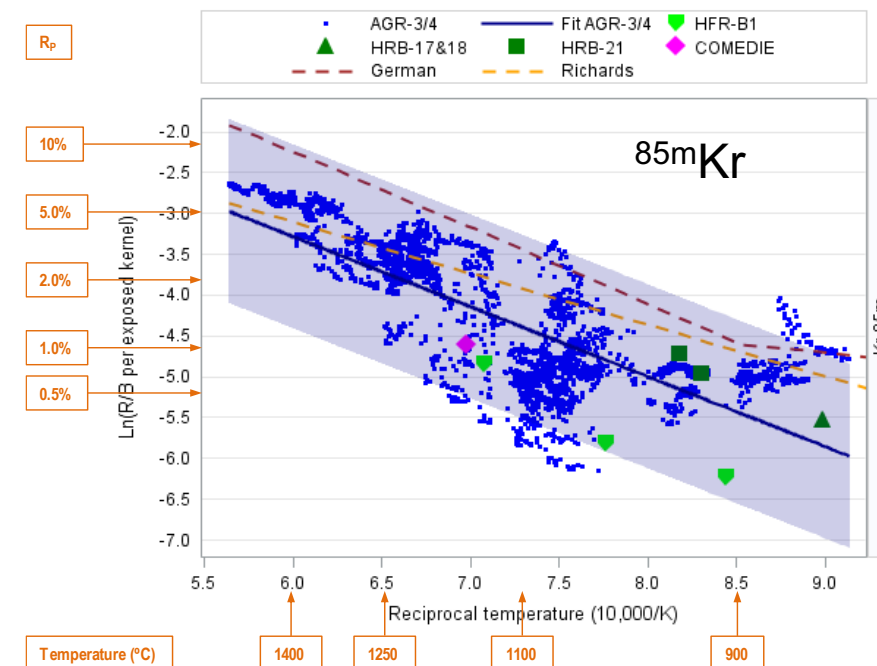
# Radionuclide Release Barriers: Particle Coatings

- Potential release mechanisms
  - Diffusion through intact coatings
  - As-fabricated coating defects
  - In-service coating failure

- Controlling parameters
  - Fuel temperatures
  - Time
  - Fast neutron fluence

- Barrier performance
  - Transport through intact coatings:
    - Ag significantly released
    - Other fission products (e.g., Sr, Eu) exhibited more modest release
  - Gases retained by OPyC with defective/failed SiC
  - Metals released when SiC fails
  - TRISO failure rates are very low in modern TRISO fuel ($\sim 5 \times 10^{-5}$ or less for normal operation)
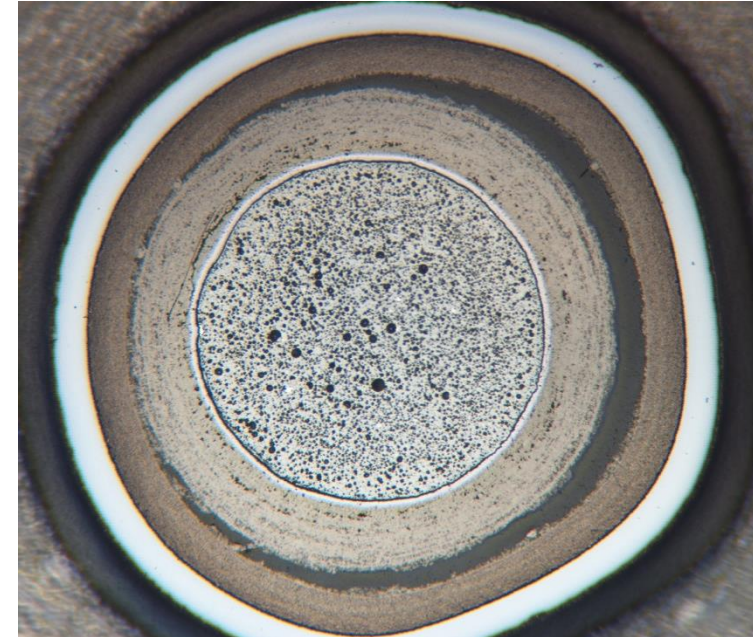
# Radionuclide Release Barriers: Matrix/graphite

- Potential release mechanisms
  - Diffusion/vaporization
  - Matrix/graphite oxidation
- Controlling parameters
  - Temperature
  - Time
  - Fast neutron fluence
  - Oxidant concentration
- Barrier performance
  - Sr and Eu exhibit fairly high retention
  - Cs and Ag exhibit lower levels of retention
  - Kr, Xe, and I not retained
  - Sorbed metals assumed to be released by oxidation

# Radionuclide Release Barriers: Primary Coolant Circuit

- Potential release mechanisms
  - Primary coolant leaks
  - Liftoff (mechanical re-entrainment)
  - Primary coolant pressure relief
  - Steam-induced vaporization
  - Washoff (removal by liquid $H_2O$)

- Controlling parameters
  - Temperatures in primary circuit
  - Size/location of coolant leaks
  - Particulate matter in primary circuit
  - Steam/liquid $H_2O$ ingress and egress

- Barrier performance
  - Condensable radionuclides plate out during normal operation
  - Circulating Kr, Xe and H-3 limited by He purification system
  - Plateout largely retained during rapid blowdowns

# Radionuclide Release Barriers: Reactor Building

- Potential release mechanisms
  - Venting through louvers
  - Building leakage
- Controlling parameters
  - Leak path(s) and rates
  - Contaminated steam/liquid $H_2O$
  - Contaminated particulate matter
  - Temperatures along leak path(s)

- Barrier performance
  - Noble gases decay during holdup
  - Condensable fission products, including iodine, deposit
  - Contaminated steam condenses
  - Contaminated dust settles out and deposits

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# Outline

- Barriers to radionuclide (RN) release in high-temperature gas-cooled reactors (HTGRs)

- Radionuclide design criteria

- Computational tools to predict radionuclide release and existing data

- Approach to estimate source term from HTGRs

# Radionuclide Design Criteria

- "Top down" approach used to determine allowable radionuclide releases within the functional containment system

- Start with imposed requirements (e.g., site-boundary dose limits)

- Allowable radionuclide inventories in primary circuit derived from radionuclide control requirements
  - Two-tier set of "Radionuclide design criteria" defined to explicitly include safety factors in plant design

# Design Margins (Safety Factors) Are Explicitly Included in Radionuclide Design Criteria (Prismatic Example)



Typical Design Margins:
4x for Fission Gases
10x for Fission Metals

A Review of Radionuclide Release from HTGR Cores During Normal Operation, EPRI report 1009382 (2003)

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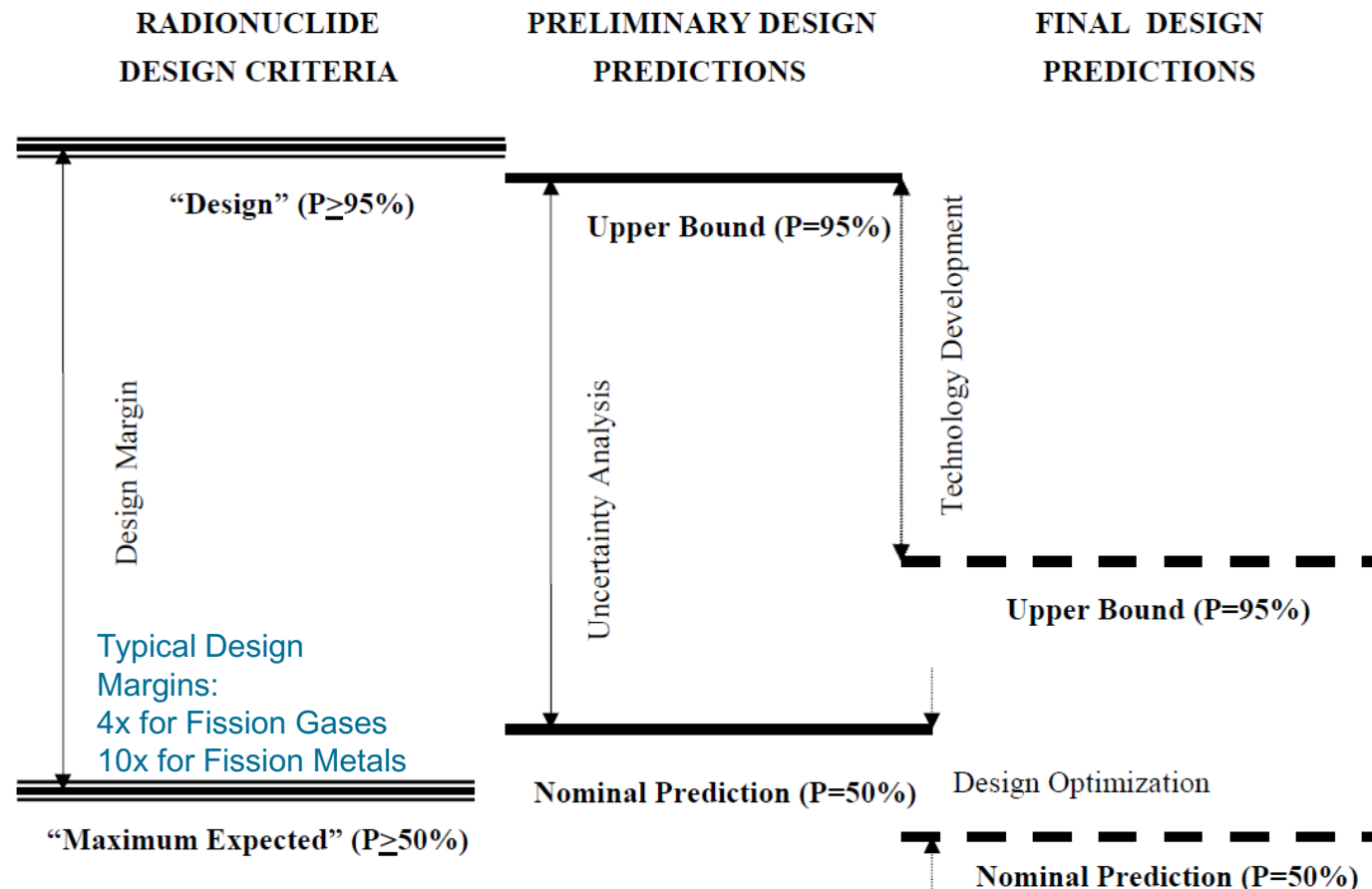
# Radionuclide Release Tradeoffs

- Tradeoffs exist between the relative allocations of the performance of some of the barriers

*Fuel quality*:
- Heavy metal contamination
- Defective particles

*Fuel performance*:
- In-service failures

*Reactor design considerations*:
- Core operating conditions
- He pressure boundary
- Reactor building

- Example: allowable fuel contamination levels may be higher if in-service failures are much lower than expected

# Outline

- Barriers to radionuclide (RN) release in high-temperature gas-cooled reactors (HTGRs)

- Radionuclide Design criteria

- Computational tools to predict radionuclide release and existing data
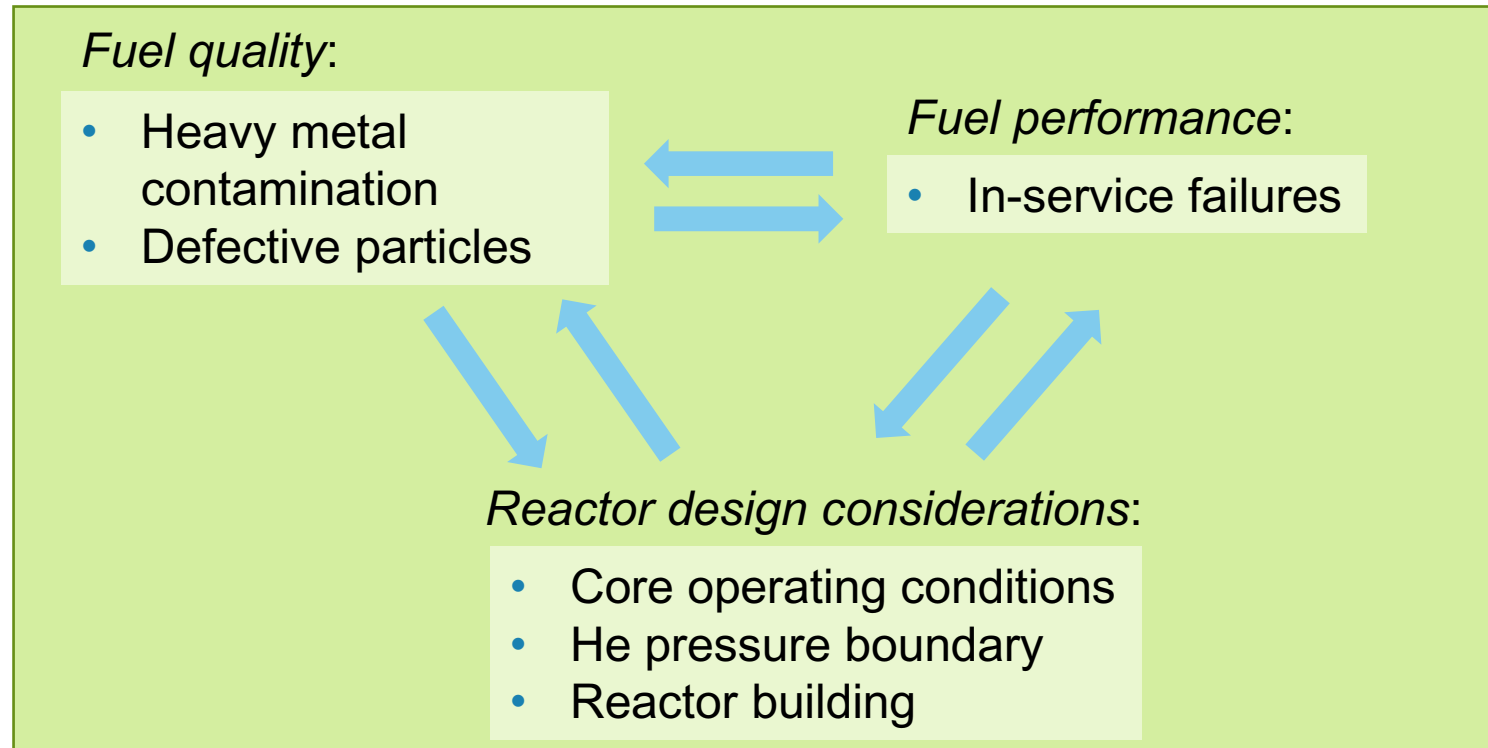
- Approach to estimate source term from HTGRs

# Computational Tools to Predict Radionuclide Behavior in HTGRs

- Design methods for predicting FP transport in HTGRs derived from experimental data
  - Typically, design codes model multiple radionuclide release barriers
  - Core analysis codes typically model fuel performance as well
  - Core codes are typically design specific (i.e., prismatic or pebble)
  - Phenomenological component models derived from data
  - Material property data (e.g., diffusivities, etc.) required as input

- Many comparisons of code predictions with experimental data
  - Reactor surveillance, in-pile tests, etc.
  - Agreement between predictions and measurements has been reasonably good, with predictions somewhat conservative relative to measurements
  - Codes not completely verified and validated
  - Comparisons are often with data from integral release tests; validation of contributions from specific mechanisms is difficult.

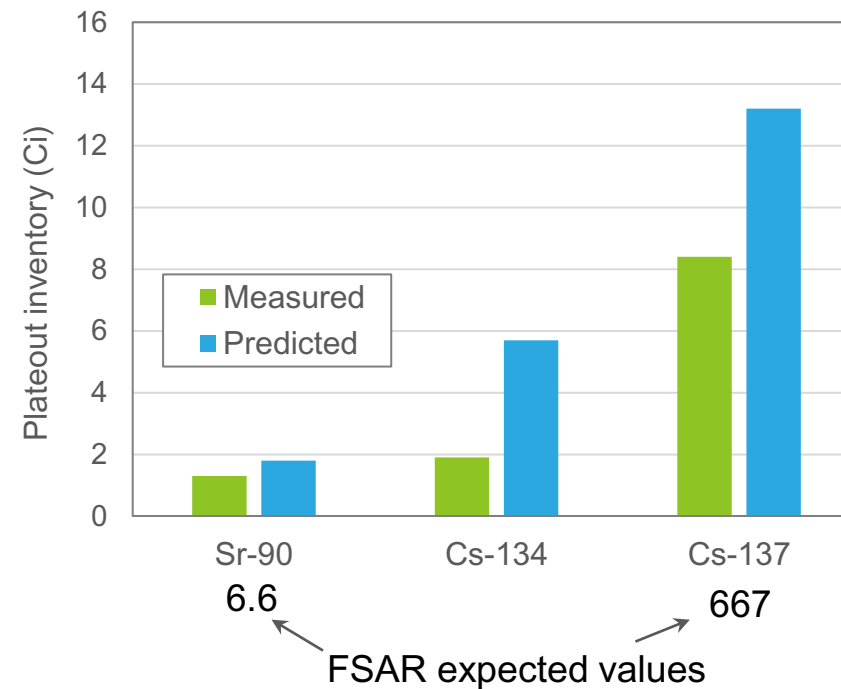# Comparison of Code Predictions with Data

- Past radionuclide release predictions have been reasonably accurate and conservative

Comparison of FSV predicted and measured Kr-85m release
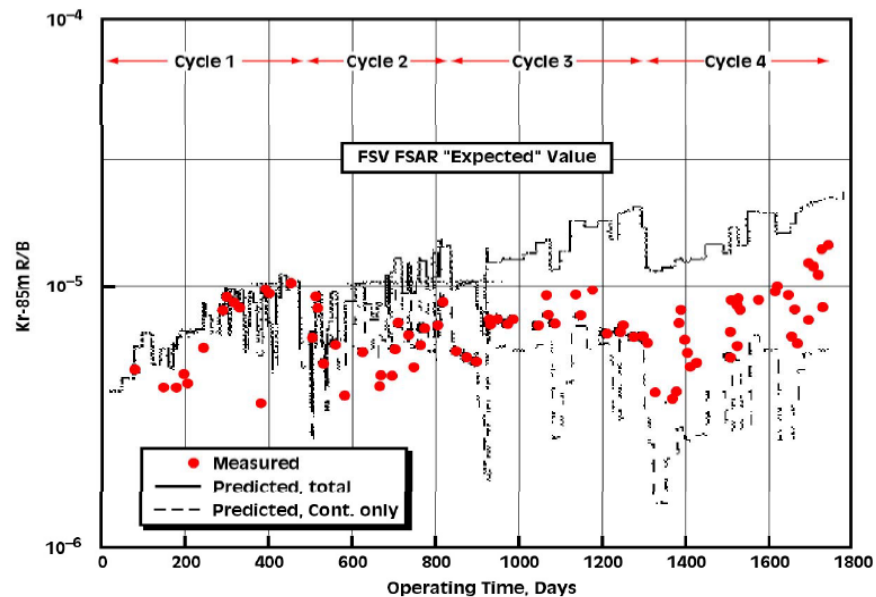


Comparison of FSV predicted and measured fission metal release



Many additional examples presented in TECDOC-978:
*Fuel performance and fission product behavior in gas cooled reactors*, IAEA, TECDOC-978 (1997)

# Dedicated Experiments to Refine Source Term Input Parameters

- US Advanced Gas Reactor (AGR) Program AGR-3/4 Irradiation Experiment

- Measure fission product transport in matrix and graphite materials
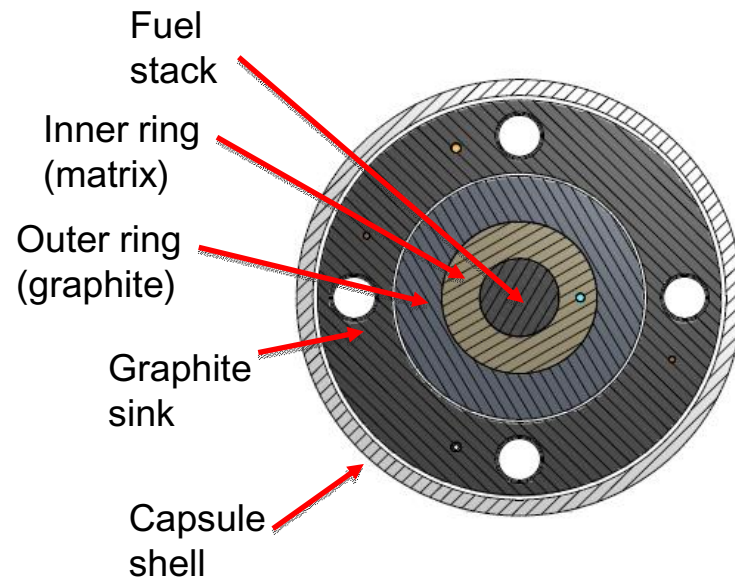


*AGR-3/4 Capsule Cross Section*

Comparison of measured and predicted Cs profile in AGR-3/4 graphite rings
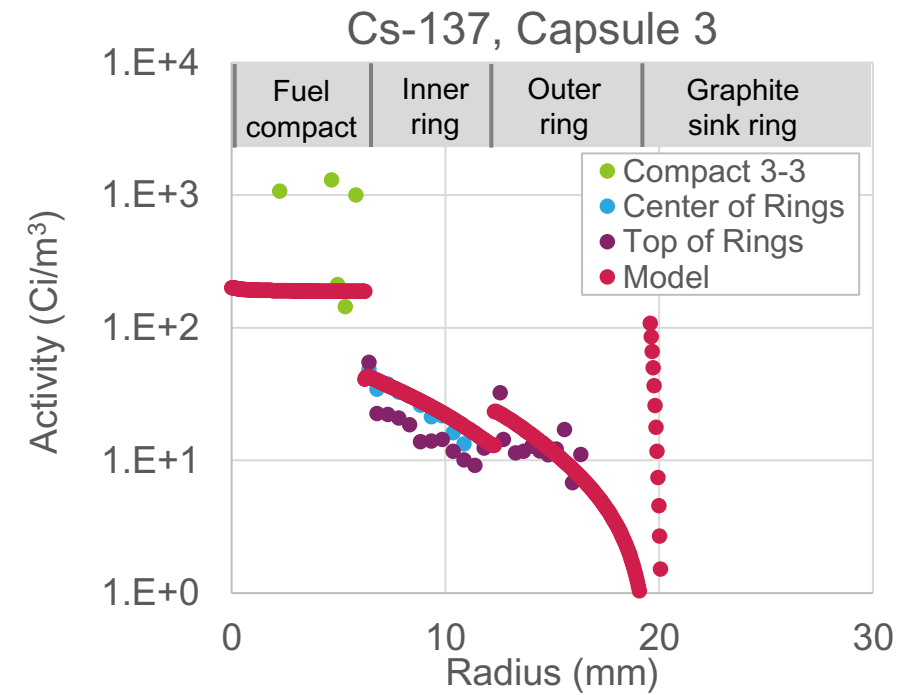
# Particulate Matter ("Dust") in Primary Circuit May Alter Fission Product Transport Behavior

- Potential sources of dust in HTGRs
  - Foreign material from initial construction or refueling
  - Abrasion/attrition of spherical fuel elements (pebble bed)
  - Erosion or corrosion of fuel or reflector blocks (prismatic)
  - Foreign material from interfacing systems (e.g., HPS)
  - Spallation of friable metallic surface films
  - Carbon deposition from CO decomposition

- Potential impact on fission product (FP) transport
  - Altered FP plateout distributions in primary circuit
  - Enhanced FP release from primary circuit into reactor building
  - Altered FP transport behavior in reactor building

- Experience:
  - Prismatic: very little dust formation; minor impact (PB1, FSV, HTTR)
  - Pebble bed: Measured and characterized in AVR; had impact on plant D & D

# Tritium Release in HTGRs

- Tritium (H-3) will be produced by nuclear reactions
  - Ternary Fission (Yield = ~$10^{-4}$)
  - Neutron activation of impurities (He-3 in coolant; Li in graphite)
  - Neutron capture in boron control materials
- Some H-3 will accumulate in primary helium
  - Controlled by He Purification System
  - Significant sorption on core graphite
- Fraction of circulating H-3 in He will permeate through intermediate heat exchanger (IHX) and SG with potential to contaminate process gases and steam
  - Generally not a concern with 750°C outlet temperatures; becomes more important at outlet of 900°C

- H-3 will contribute to public and occupational exposures
  - Environmental releases from plant (liquid discharge)
  - Contaminated products (e.g., hydrogen, bitumen)
- Data on H-3 transport in reactors and in relevant materials have been obtained with dedicated experiments and through reactor operating experience
- In operating reactors, offsite H-3 release has been below regulatory limits

# HTGR RN Transport Knowledge Base

- Extensive international data base on HTGR fission product  transport available to support HTGR design & licensing
  - Summarized in IAEA TECDOC-978 (1997)
  - IAEA TECDOC-1645 (2010) – Summary of fission product transport modeling approaches
- Primary data sources
  - Previous HTGR R&D programs in USA, FRG, Japan, France, etc.
  - Reactor surveillance programs (seven HTGRs constructed)
  - On-going R&D programs, especially AGR fuel program
- Existing data base has limitations; hence, uncertainties in  models and material properties are often large
  - Some data are for non-reference materials
  - QA pedigree uncertain
- Additional testing needed to complete validation of design  methods for predicting fission product source terms

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# Approach to Estimate Source Terms and Demonstrate the Impact of Barrier Performance

- D.A. Petti et al., *Representative source terms and the influence of reactor attributes on functional containment in modular high-temperature gas-cooled reactors*, Nucl. Tech. 184 (2013) 181-197

- Radionuclide release from core during normal operation from 4 sources:
  - Release from heavy metal contamination
  - Release from TRISO fuel with SiC defects
  - Release from in-service particle failures
  - Diffusive release through fuel particle coatings

- Total radionuclide inventory in the fuel calculated (e.g., ORIGEN)

- "Attenuation factors" (AFs) are applied at various levels to account for retention of radionuclides (calculated at 50% and 95% confidence)
  - Kernels
  - Coatings
  - Graphite
  - He pressure boundary (liftoff)
  - Reactor building

- AFs determined based on expert opinion (informed by experimental data, calculations)

- Accommodate uncertainties in defect/failure fractions and AFs using Monte Carlo approach

# mHTGR Source Term During Normal Operation
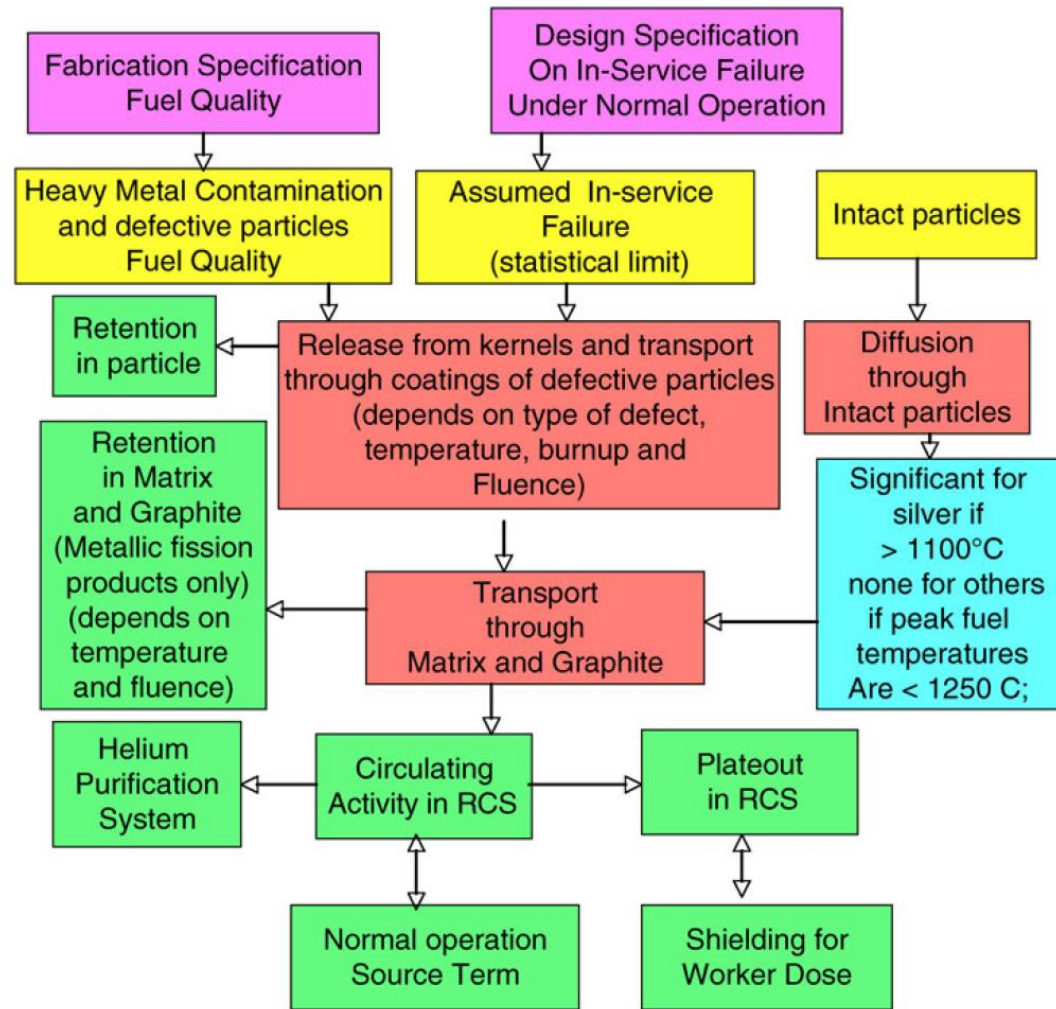
# Calculating Inventory Released from Core from Separate Sources

**Release from *heavy metal contamination* (includes contamination outside SiC layers and exposed kernels)**

$$R^i_{HMC} = \frac{Inv^i * HMC}{AF^i_{HMC} * AF^i_G}$$

**Release from particles with defective SiC and in-service failure**

$$R^i_{DSiC+ISF} = \frac{Inv^i * (DSiC + ISF)}{AF^i_K * AF^i_G}$$

$HMC$ = level of heavy metal contamination

$Inv^i$ = inventory of fission product $i$

$R^i_{HMC}$ = release of fission product $i$ from HMC (Ci)

$R^i_{DSiC+ISF}$ = release of fission product $i$ from SiC defects and in-service failures (Ci)

$AF^i_{HMC}$ = attenuation factor of fission product $i$ for HMC

$AF^i_G$ = attenuation factor of fission product $i$ in graphite

$AF^i_K$ = attenuation factor of fission product $i$ in kernel

$DSiC$ = level of SiC defects

$ISF$ = level of in-service failures

- Similar equations for release through intact particles, inventory retained in the graphite, and plate-out inventory
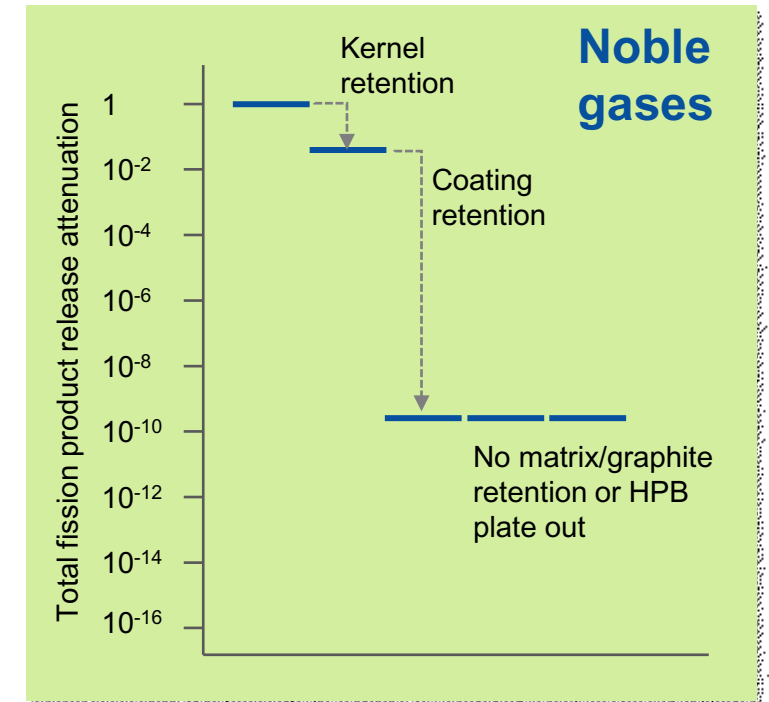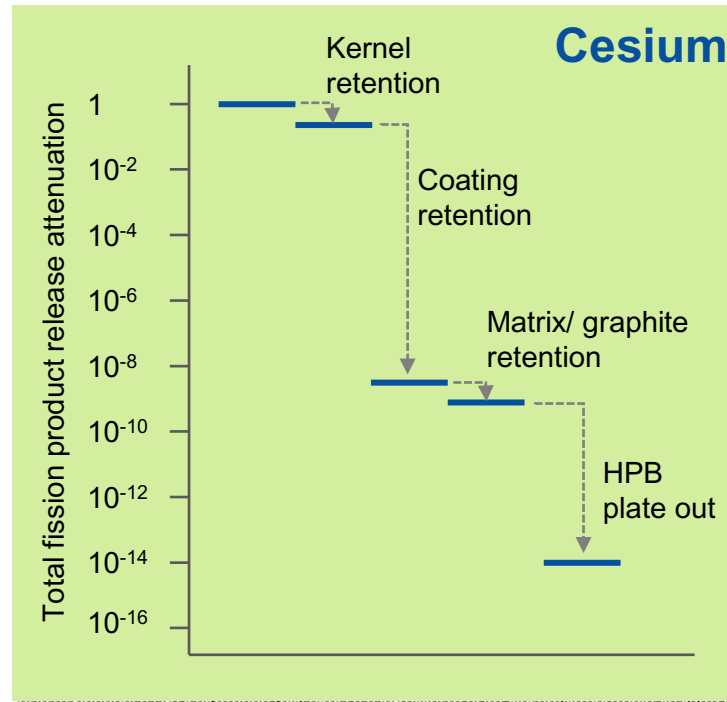
# Radionuclide Release Attenuation Factors

Prismatic reactor barrier core-average attenuation factors
during normal operations (700°C Reactor Outlet Temperature)

| Fission Product Class | Heavy Metal Contamination | | Fuel Particle Kernel | | Diffusive Release through Fuel Particle Coatings | | Graphite (Compact Matrix and Fuel Element) | | Helium Pressure Boundary | |
|---|---|---|---|---|---|---|---|---|---|---|
| Confidence Limit | $AF_{HMC}$ 50% | $AF_{HMC}$ 95% | $AF_K$ 50% | $AF_K$ 95% | $AF_{Diff}$ 50% | $AF_{Diff}$ 95% | $AF_G$ 50% | $AF_G$ 95% | $AF_{HPB}$ 50% | $AF_{HPB}$ 95% |
| Noble Gases | 10 | 3 | 50 | 17 | $10^8$ | $10^7$ | 1 | 1 | 1 | 1 |
| I, Br, Se, Te | 10 | 3 | 50 | 17 | $10^8$ | $10^7$ | 1 | 1 | $10^6$ | $10^5$ |
| Cs, Rb | 1 | 1 | 3 | 1 | $10^8$ | $10^6$ | 5 | 2 | $10^6$ | $10^5$ |
| Sr, Ba, Eu | 1 | 1 | 50 | 20 | $10^3$ | 200 | $10^3$ | 300 | $10^6$ | $10^5$ |
| Ag, Pd | 1 | 1 | 2 | 1 | 500[a] | 100[a] | 2 | 1 | $10^6$ | $10^5$ |
| Sb | 1 | 1 | 2 | 1 | $10^8$ | $10^6$ | 20 | 2 | $10^6$ | $10^5$ |
| Mo, Ru, Rh, Tc | 1 | 1 | 500 | 30 | $10^8$ | $10^7$ | $10^3$ | 300 | $10^6$ | $10^5$ |
| La, Ce | 1 | 1 | 500 | 30 | $10^8$ | $10^7$ | $10^3$ | 300 | $10^6$ | $10^5$ |
| Pu, actinides | 1 | 1 | $10^3$ | 100 | $10^8$ | $10^7$ | $10^4$ | $10^3$ | $10^6$ | $10^5$ |

[a] Values presented here for Ag-110m. For Ag-111, the values for the diffusive release through the coating are increased by a factor of 5 to account for the effect of the half-life on the release.

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# Radionuclide Barrier Performance

- Examples of multiple barrier performance:
  - Radionuclide release from *intact particles* to helium pressure boundary during normal operation
  - Prismatic HTGR 700°C outlet temperature



**Cesium**

Total fission product release attenuation

Kernel retention

Coating retention

Matrix/ graphite retention

HPB plate out

**Noble gases**

Total fission product release attenuation

Kernel retention

Coating retention

No matrix/graphite retention or HPB plate out

# Radionuclide Inventories During Normal Operation: Representative Examples for Prismatic Reactor Design

Mean values for I-131, Cs-137, and Sr-90 inventories (curies) released to the helium pressure boundary and retained in the fuel matrix and graphite

| Reactor Design Configuration | I-131 | | Cs-137 | | Sr-90 | |
|---|---|---|---|---|---|---|
| | In Fuel Matrix and Graphite | In He Pressure Boundary | In Fuel Matrix and Graphite | In He Pressure Boundary | In Fuel Matrix and Graphite | In He Pressure Boundary |
| 600 MW(t) Prismatic 700°C ROT | nil | 30 | 24 | 5 | 2750 | 0.1 |
| 600 MW(t) Prismatic 900°C ROT | nil | 74 | 226 | 254 | 5680 | 31 |

*ROT: Reactor outlet temperature*

D.A. Petti et al., *Representative source terms and the influence of reactor attributes on functional containment in modular high-temperature gas-cooled reactors*, Nucl. Tech. 184 (2013) 181-197

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# Evaluating Radionuclide Release During Licensing Basis Events

- Similar approach taken for accident scenarios to determine total source term released from reactor building

- Radionuclide sources in accidents
  - Release from heavy metal contamination
  - Release from TRISO fuel with SiC defects
  - Release from in-service particle failures
  - Diffusive release through fuel particle coatings
  - Inventory in the graphite/matrix from normal operation
  - Lift-off of inventory plated out on the coolant boundary during normal operation

- Different accident scenarios have specific attenuation factors depending on accident conditions (temperature, dry/wet conditions, etc.)
  - e.g., moisture can increase release from exposed kernels and lift-off from the pressure boundary

- Separate calculations for short- and long-term release for accidents (driven by differing half-lives of radioisotopes)

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# Evaluating Source Term for Licensing Basis Events
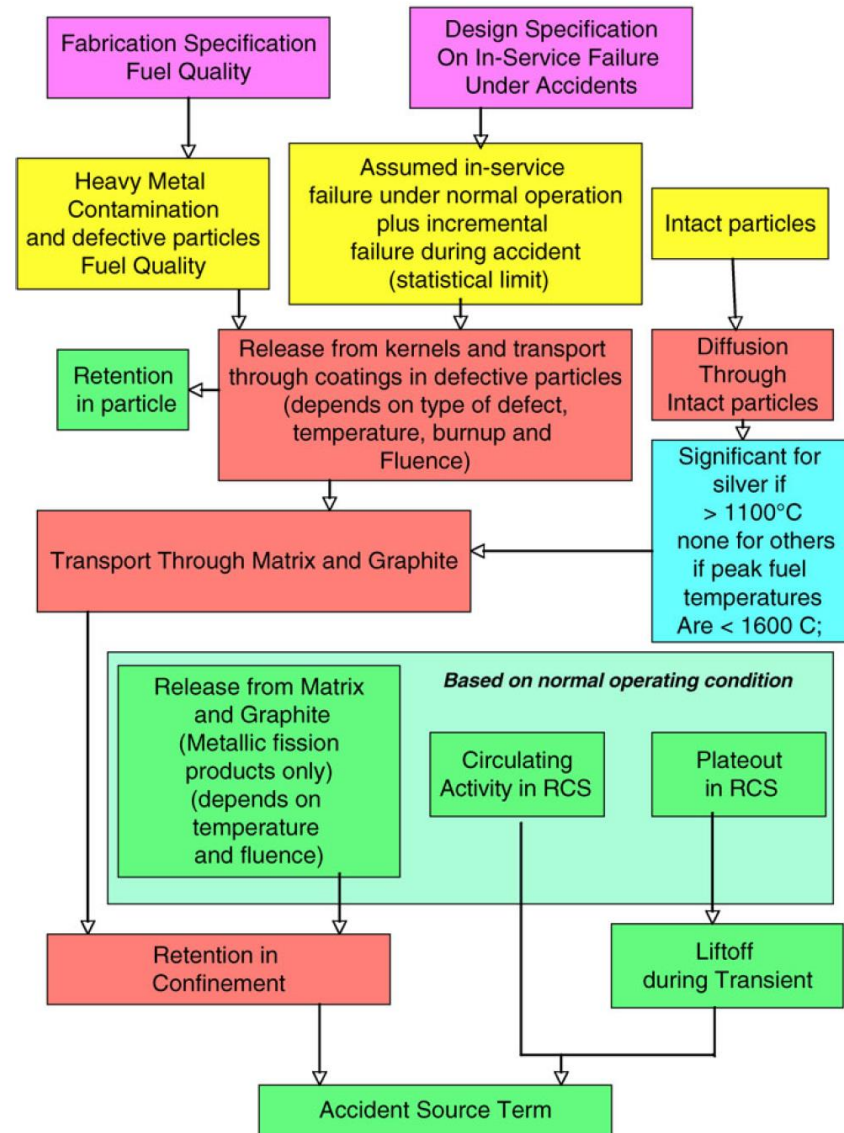


D.A. Petti et al., *Representative source terms and the influence of reactor attributes on functional containment in modular high-temperature gas-cooled reactors*, Nucl. Tech. 184 (2013) 181-197
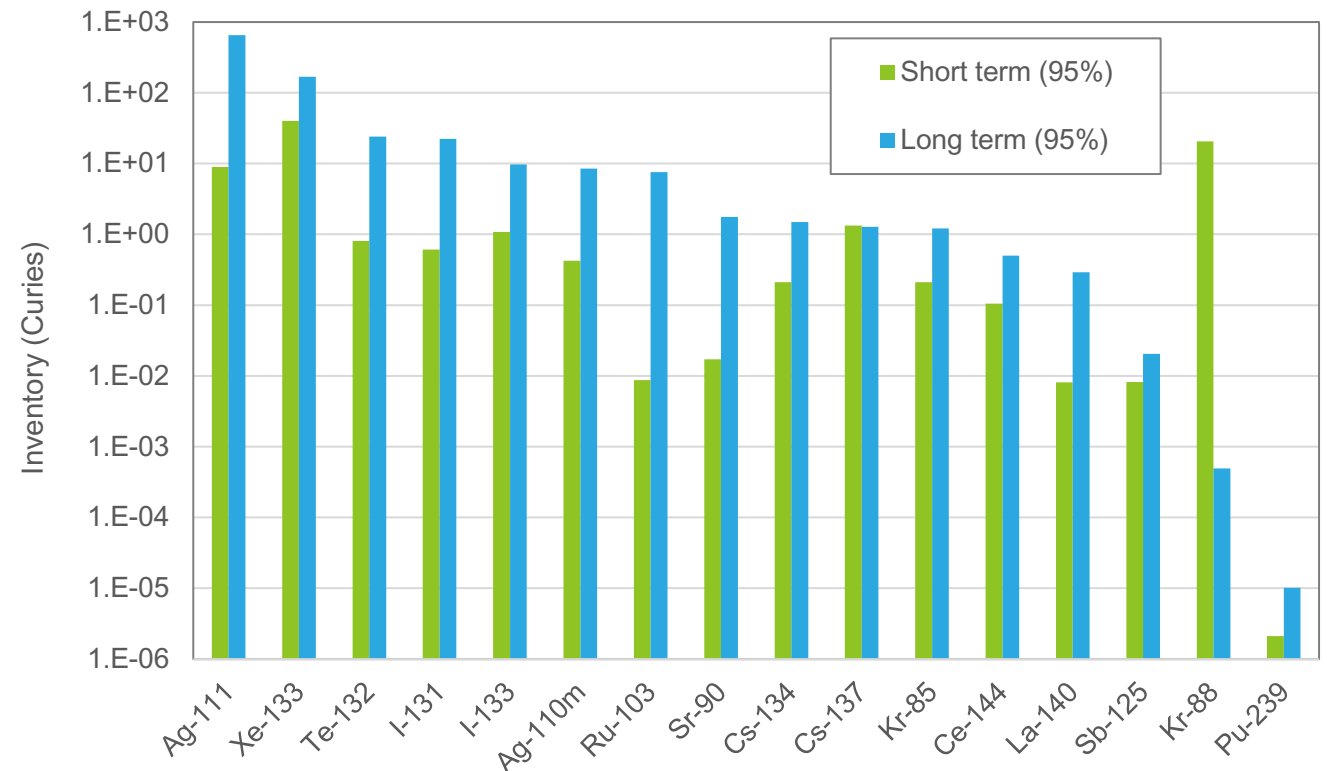
# Radionuclide Release Attenuation Factors During Accidents

700°C and 900°C ROT long-term accident release and attenuation factors for a break in helium pressure boundary

| Fission Product Class | Accident Release | | Attenuation Factors | | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|---|---|---|
| | Incremental Failure- Accident | | Heavy Metal Accident Release | | Non-Intact Accident Release | | Intact Accident Release | | Graphite | | Helium Pressure Boundary | | Reactor Building | |
| Confidence Limit | 50% | 95% | AF 50% | AF 95% | AF 50% | AF 95% | AF 50% | AF 95% | AF 50% | AF 95% | AF 50% | AF 95% | AF 50% | AF 95% |
| Noble Gases | $3\times10^{-5}$ | $8\times10^{-5}$ | 2 | 1 | 10 | 5 | $10^7$ | $2\times10^6$ | 1 | 1 | 5 | 2 | 1 | 1 |
| I, Br, Se, Te | $3\times10^{-5}$ | $8\times10^{-5}$ | 2 | 1 | 10 | 5 | $10^7$ | $2\times10^6$ | 1 | 1 | 5 | 2 | 10 | 2 |
| Cs, Rb | $3\times10^{-5}$ | $8\times10^{-5}$ | 1 | 1 | 1 | 1 | $10^7$ | $2\times10^6$ | 10 | 3 | 10 | 3 | 10 | 2 |
| Sr, Ba, Eu | $3\times10^{-5}$ | $8\times10^{-5}$ | 1 | 1 | 1 | 1 | $10^6$ | $2\times10^5$ | 100 | 30 | 10 | 3 | 10 | 2 |
| Ag, Pd | $3\times10^{-5}$ | $8\times10^{-5}$ | 1 | 1 | 1 | 1 | 500 | 100 | 1 | 1 | 10 | 3 | 10 | 2 |
| Sb | $3\times10^{-5}$ | $8\times10^{-5}$ | 1 | 1 | 1 | 1 | $10^6$ | $2\times10^5$ | 100 | 30 | 10 | 3 | 10 | 2 |
| Mo, Ru, Rh, Tc | $3\times10^{-5}$ | $8\times10^{-5}$ | 1 | 1 | 100 | 50 | $10^7$ | $2\times10^6$ | 10 | 3 | 10 | 3 | 10 | 2 |
| La, Ce | $3\times10^{-5}$ | $8\times10^{-5}$ | 1 | 1 | 100 | 50 | $10^7$ | $2\times10^6$ | 100 | 30 | 10 | 3 | 10 | 2 |

D.A. Petti et al., *Representative source terms and the influence of reactor attributes on functional containment in modular high-temperature gas-cooled reactors*, Nucl. Tech. 184 (2013) 181-197

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# Radionuclide Source Terms During LBEs: Representative Example for Prismatic Reactor Design

Source terms for a break in He pressure boundary, upper 95% confidence levels 600 MW(t) Prismatic 700°C reactor outlet temperature (Table X)



D.A. Petti et al., *Representative source terms and the influence of reactor attributes on functional containment in modular high-temperature gas-cooled reactors*, Nucl. Tech. 184 (2013) 181-197

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# Summary

- HTGR designs employ multiple radionuclide release barriers to meet radionuclide control requirements

- Radionuclide transport in HTGRs has been extensively investigated

- Design methods available to predict performance of the radionuclide release barriers during normal operation and accidents
  - Codes have been used extensively for reactor design and analysis, including operating HTGRs

- Many comparisons of code predictions with data
  - Reactor surveillance, in-pile tests, etc.
  - Comparisons are often with data from integral release tests; validation of contributions from specific mechanisms is difficult
  - Codes not completely verified and validated

- Additional data from ongoing programs (e.g., US DOE AGR program) will help refine transport parameters and reduce uncertainties

- A proposed approach involves estimating radionuclide attenuation at each barrier based on empirical data and calculations to bound release behavior

# Suggested Reading

- 2010 HTGR Technology Course for the Nuclear Regulatory Commission

- A Review of Radionuclide Release from HTGR Cores During Normal Operation, EPRI report 1009382 (2003)

- D.A. Petti et al., Representative source terms and the influence of reactor attributes on function containment in modular high-temperature gas-cooled reactors, Nucl. Tech. 184 (2013) 181-197

- Fuel performance and fission product behavior in gas cooled reactors, IAEA, TECDOC-978 (1997)

- High Temperature Gas Cooled Reactor Fuels and Materials, IAEA, TECDOC-1645 (2010)