HTGR Technology Course for the Nuclear Regulatory Commission

May 24 – 27, 2010

Module 13 Fission Product Behavior in HTGRs







Outline

- Introduction, background, and radionuclide fundamentals
- Radionuclide (RN) transport in HTGRs
 - Fuel kernels
 - Particle coatings
 - Fuel matrix/graphite
 - Primary coolant circuit
 - Reactor building
- Design methods for predicting RN transport
- Comparison of code predictions with data
 - In-pile test data
 - Reactor surveillance data





Introduction and Background

- HTGR designs employ multiple radionuclide (RN) release barriers to meet RN control requirements
- RN transport in HTGRs has been extensively investigated
- Design methods available to predict performance of the RN release barriers during normal operation and accidents
 - Codes have been used extensively for reactor design & analysis, including operating HTGRs
- Many comparisons of code predictions with data
 - Reactor surveillance, in-pile tests, etc.
 - Codes not completely verified and validated
- NGNP/AGR Fuel Program will complete validation of codes
 - Single-effects data for component model upgrades
 - Independent integral data for code validation





Radionuclide Containment Function

- HTGR designs employ multiple RN release barriers to meet RN control requirements
 - Fuel kernels
 - **<u>Particle coatings</u>** (most important barrier)
 - Fuel-element matrix/fuel-element graphite (prismatic)
 - Primary coolant pressure boundary
 - Reactor building (RB)
- These multiple RN barriers provide Defense-in-Depth
- Performance criteria for each RN release barrier derived using a top-down allocation process (Module 3)





HTGR RN Sources and Pathways



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HTGR Radionuclide Inventories

- Radionuclide inventories in reactor core calculated using standard burnup/depletion codes; for example:
 - ORIGEN for core RN inventories
 - GARGOYLE (GA)/VSOP99 (PBMR) for decay heat calculations
- Allowable RN inventories in primary circuit derived from RN control requirements using top-down functional analysis
 - Two-tier set of "RN design criteria" defined to explicitly include safety factors in plant design (next slide)
 - RN inventories specified for:
 - Circulating activity in primary coolant
 - Plateout activity in primary circuit
 - He purification system
 - Plant mass balance calculated with RADC (GA)/DAMD (PBMR)





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



Radionuclide Release Fundamentals

$(f.r.)_{core} = \frac{C(f.r.)_{c} + F(f.r.)_{F} + [1 - C - F](f.r.)_{D}}{AF_{graphite}}$

- (f.r.)_{core} = fractional release from core
- C = heavy-metal contamination fraction
- $(f.r.)_{C}$ = fractional release from contamination
- F = failure fraction:
- $(f.r.)_{F}$ = fractional release from failed particles
- (f.r.)_D = fractional diffusive release from intact particles
- AF_{graphite} = matrix/graphite attenuation factor





Dominant Radionuclides in HTGRs

Nuclide	Half Life	Primary Impact			
I-131	8 day	Offsite dose, O&M dose			
Ag-110m	250 day	O&M dose			
Cs-137	30 yr	O&M dose, offsite dose			
Cs-134	2.1 yr	O&M dose, offsite dose			
Sr-90	28 yr	Offsite dose			
Kr & Xe		Normal operation gaseous effluent			
H-3	12.3 yr	Normal operation liquid effluent; product contamination			



Significance of Tritium for HTGRs

Tritium (H-3) will be produced in NGNP by nuclear reactions

- Ternary Fission (Yield = $\sim 1 \times 10^{-4}$)
- 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 IHX & SG with potential to contaminate process gases and steam
- H-3 will contribute to public & occupational exposures
 - Environmental releases from plant (liquid discharge)
 - Contaminated products (e.g., hydrogen, bitumen, etc.)



H-3 Permeation through Metals Suppressed by Oxide Surface Films



- H-3 Permeation Measurements at ORNL
 - ✓ Incoloy 800 steam generator tubing
 - ✓ Trace H-3 in He on outside of tube
 - \checkmark Steam on inside
- H-3 permeation behavior
 - ✓ Rapid decrease during first 36 hr
 - ✓ Subsequent slow decrease
 - ✓ Square-root time dependence
 - ✓ Growing oxide layer on steam side
 - H-3 permeation through oxide layer rate limiting
- Implications for steam-cycle HTGRs
 - \checkmark He and H₂O chemistry important
 - ✓ Oxide layers inhibit H-3 permeation
 - Thermal cycling reduces effectiveness of oxide layer (cracking)





H-3 Behavior in Prismatic HTGRs Off-Site H-3 Releases < Regulatory Limits

Dragon HTR

- First determination of H-3 behavior in an HTGR
- Li_2SO_4 (neutron poison) in secondary H_2O unique source
- H_2O injection increased circulating H-3

Peach Bottom 1

- ~60% H-3 release from BISO fuel; retained in graphite
- H-3 produced in control materials retained in place
- Small H-3 permeation into secondary coolant (~1.1 Ci)

• Fort St. Vrain (FSV)

- Ti getters in HPS did not meet requirements (deactivated by N₂,etc.)
- Core graphite major sink for H-3: >10x more effective than HPS
- Frequent H_2O Ingresses released H-3 from core graphite
- HTTR
 - H-3 plant mass balance at 938°C core outlet temperature
 - Extensive data on H-3 permeation through IHX (Hasteloy XR)



FSV Tritium Concentration in Primary Helium



H-3 Behavior in Pebble-Bed HTGRs Off-Site H-3 Releases < Regulatory Limits

• AVR

- High lithium content in "carbon brick" side reflector (~4 ppm)
 - Dominant source of H-3 production
 - <50 ppb typical for HTGRs</p>
- H_2 (protium) injection tests
 - Displaced H-3 sorbed on core graphite
 - Decreased H-3 permeation to secondary H_2O
- Adjusted feed-water chemistry promoted growth of oxide layer on SG tubes reducing H-3 permeation and release
- Large SG leak resulted in large H-3 release from core graphite

• THTR

- Little published information
- HTR-10
 - No published information





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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
- 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 fuel AGR 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





Radionuclide Release Barriers Fuel Kernels

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 > Sr)
- Other radionuclides, including actinides, completely retained







Fission Gas Release Fundamentals Chemical Element and Half-Life Dependence



1. Release Rate-to-Birth Rate Ratio (R/B) $\alpha \; \sqrt{t_{1/2}}$

3.
$$R/B_{Br, Se} = R/B_{Kr}$$

 $R/B_{I,Te} = R/B_{Xe}$

4. Behavior predicted by Booth Eqn

$$\left(\frac{R_{B}}{B} \right)_{ji} = 3 \sqrt{\left(\frac{\xi_{j}}{\lambda_{i}} \right)} ,$$

5. Deviations observed, especially at lower temperatures and high neutron fluxes (e.g., in HFIR)





Fission Gas Release from Failed Particles Effect of H₂O on Kr-85m R/B



increased steady-state **R/B upon hydrolysis** Stage 2. increased steady-state R/B Stage 3. decline to

nearly initial R/B upon removal of H₂O

Pattern repeats with multiple H₂O injections

Effect of H₂O on Kr-85m R/B from failed UCO particles at 755°C in HRB-17 Test (Original UCO kernel had been converted to UO_2 by previous H_2O injection)





Dependence of Fission Gas Release on H₂O Partial Pressure



- Previous plot showed R/B vs. time after H₂O injection
 - ✓ Instantaneous release rate/birth rate
 - R/B spiked because of stored gas release upon hydrolysis
- Cumulative release divided by cumulative birth (FR) shown here
- FR dependence on H₂O partial pressures
 - \checkmark Linear increase at low partial pressures
 - ✓ Independent at high partial pressures
- Typical behavior for gas-solid reactions
 - ✓ e.g., "Langmuir-Hinshelwood" kinetics
- Significant for large H₂O ingress accidents
 - ✓ FR reaches plateau at ~20% becoming independent of H₂O partial pressure



Fission Product Release from LEU UO₂ Kernels under Core Conduction Cooldown Conditions



Heating Time (hr)

- Postirradiation heating of FGR LEU UO₂ bare kernels from FRJ2-P28/C6
- Test articles reactivated prior to heating to generate short-lived radionuclides (e.g., I-131)
- FP release behavior as temperature ramped from 1000 to 1600°C:
 - ✓ Xe-133, I-131 ("J-131") and Ag-110m rapidly released
 - ✓ Cs-137 delayed but reaches 100%
 - Sr-90 substantially retained for long times
- Kernel release rates expected to increase at higher burnups (low-burnup ThO₂ data)





Radionuclide Release Barriers Particle Coatings

• Potential release mechanisms

- Diffusion through intact coatings
- In-service coating failure
- SiC corrosion by fission products
- SiC thermal decomposition

Controlling parameters

- Fuel temperatures
- Time
- Fast neutron fluence (Increased FP diffusivities)
- **Barrier performance** (Module 8)
 - Only Ag (and H-3) released by diffusion from intact particles
 - No pressure-induced failure of standard particles
 - SiC thermochemical failure function of time/temperature
 - Gases retained by OPyC with defective/failed SiC









Fission Product Release from LEU UO₂ TRISO Particles under Core Conduction Cooldown Conditions



- Postirradiation heating of German LEU UO₂ TRISO particles in spheres at 1600 & 1800°C
- No complete coating failure (1 particle failure would yield Kr-85 fractional release = ~10⁻⁴
- FP release at 1600°C
 - ✓ Rapid Ag-110m release
 - ✓ Kr-85, Cs-137, and Sr-90 completely retained
- FP release at 1800°C
 - ✓ Kr-85, Cs-137, and Sr-90 release increasing
 - ✓ Evidence of SiC degradation (expected)
- FP transport in SiC in such tests is ambiguous
 - ✓ Degradation of SiC @ T > ~1600°C
 - ✓ FP retention, especially Sr-90, in matrix
- Longer duration tests with intact particles needed to derive effective SiC diffusivities

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FP Transport in Coatings Has Been Characterized



- HRB-22 UO₂ LEU fuel (Japanese fuel) heated at ORNL
- 25 irradiated fuel particles recovered from fuel compact
- Isothermal heating at 1700°C
- Low fractional release of Kr-85 indicates no complete coating failure (1 failed particle = 4% release)
- Some SiC degradation may have occurred at 1700°C
- Derived fission metal diffusivities in SiC coating conservative if SiC degraded

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📌 GENERAL ATOMICS

Metal Diffusivities in SiC Coatings







Radionuclide Release Barriers Core Matrix/Graphite

Potential release mechanisms

- Diffusion/vaporization
- Matrix/graphite oxidation
- Controlling parameters
 - Temperature
 - Time
 - Fast neutron fluence
 - H_2O Concentration
- Barrier performance





- Cs and Sr partially released at hotter locations
- Released Cs and Sr partially resorb on cooler graphite
- Sorbed metals assumed to be released by oxidation





Cesium Transport in Nuclear Graphite Has Been Characterized



Cs Diffusion in Nuclear Graphites



Cs Sorption on H-451 Graphite





Radionuclide Release Barriers Primary Circuit

• Potential release mechanisms

- Primary coolant leaks
- Liftoff (mechanical reentrainment)
- Primary coolant pressure relief
- Steam-Induced vaporization
- Washoff (removal by liquid H₂O)

• Controlling parameters

- Temperatures in primary circuit
- Size/location of coolant leaks
- Particulate matter in primary circuit
- Steam/Liquid H₂O ingress and egress

Barrier performance

- Condensable RNs plate out during normal operation
- Circulating Kr, Xe and H-3 limited by HPS
- Plateout largely retained during rapid blowdowns
- RN holdup due to thermal contraction of gas in vessel





Iodine Sorption on Low-Alloy Steel at 400 °C







Radionuclide Release Barriers Reactor Building

• Potential release mechanisms

- Venting through louvers
- Building leakage

Controlling parameters

- Leak path(s) and rates
- Contaminated steam/liquid H₂O
- Contaminated particulate matter
- Temperatures along leak path(s)

• Barrier performance

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





Data Base for FP Transport in Water-Reactor Containments Generally not Applicable to Modular HTGRs



PHEBUS FPT-1: FP Transport in PWR Containment

- Extensive international data base, including large integral tests; e.g.,
 - ✓ DEMONA
 - ✓ MARVIKEN
 - ✓ LACE
 - ✓ PHEBUS
- Generally not applicable to HTGRs
 - ✓ Wrong composition ("corium")
 - \checkmark Reactive aerosols
 - \checkmark Large mass concentrations
 - ✓ Different RB environment
- Some limited data may apply; e.g.,
 - \checkmark I partitioning in steam/liquid H₂O
- HTGR-specific data needed
 - ✓ Physical/chemical forms
 - ✓ Mass concentrations
 - Environment





German ALEX Test Program to Characterize Cs Aerosols in Large HTGRs during Core Heatup Accidents



German ALEX Test Program

Support HTR-500 (1390 MW_t pebble-bed HTGR) licensing

Simulate and characterize Cs aerosol formation during core heatup accidents ($T_{max} > 1800^{\circ}$ C)

Cs-saturated He ($P_{Cs} = 5 \times 10^{-3}$ atm) introduced into N_2 filled vessel at 1 atm pressure

Resulting aerosol, mainly CsOH, remained in sub-micron size range

Limited applicability to MHTGR with $P_{Cs} < \sim 1 \times 10^{-7}$ atm





CsOH Particle Size Distribution



Particulate Matter ("Dust") in Primary Circuit May Alter FP 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



Impact of Dust in Operating HTGRs

- Peach Bottom: carbon deposit from oil ingress; no impact
- FSV: rust from H₂O ingresses; no impact
- HTTR: dust from abrasion of graphite piston rings in purified He compressors; core insignificant source of dust
- AVR: graphite dust from pebbles; impact on plant D&D
- THTR: pebble debris from control rod insertion directly into pebble bed; broken pebbles impacted plant availability
- HTR-10: no published dust data





AVR Dust Best Characterized

Dust Particle Size Distribution





• AVR dust sampled and characterized

Transient Circulator Tests

- Coolant and surface concentrations
- Composition (graphitic, amorphous carbon)
- Particle size distribution of circulating dust
- Specific radionuclide loadings on dust
- Transient circulator tests to determine dust reentrainment potential



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Design Methods for Predicting Fission Product Transport in HTGRs

- Design methods for predicting FP transport in HTGRs derived from experimental data
 - Typically, design codes model multiple RN release barriers
 - Core analysis codes typically model fuel performance as well
 - Typically, core codes are 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. (some examples follow)
 - Codes not completely verified and validated
- NGNP/AGR Fuel Program will complete validation of codes
 - Single-effects data for component model upgrades
 - Independent integral data for code validation





Current GA Fuel/Fission Product Codes for Normal Operation

Code	Application			
SURVEY	Full-core, burnup, fast fluence, temperature, and fuel failure distributions; fission gas release			
TRAFIC	Full-core fission metal release			
PADLOC	Plateout distributions in primary circuit			
RADC	Overall plant mass balance for radionuclides (Advanced RANDI code available)			
TRITGO	Overall plant mass balance code for tritium (new H-3 mass balance code may be written)			



Current GA Fuel/Fission Product Codes for Accident Conditions

Code	Application			
SORS	Transient, full-core, fuel failure and fission product release (gases and metals)			
OXIDE	Transient, full-core, graphite corrosion and fuel hydrolysis for large H ₂ O ingress			
POLO	Transient FP release from primary circuit into reactor building; FP transport in RB (SANDIA developing HTGR version of MELCOR)			
MACCS	Fission product transport in environment and radiological doses (SANDIA developing HTGR version of MELCOR)			



Current PBMR Fuel/Fission Product Codes for Normal Operation

Code	Application		
VSOP99	Neutronics, fuel and graphite temperatures		
NOBLEG	Fission gas release		
FIPREX/GETTER	Fission metal release		
DAMD	Plateout and dust distributions in primary circuit; overall plant mass balance for radionuclides		



Current PBMR Fuel/Fission Product Codes for Accident Conditions

Code	Application			
TINTE	Transient graphite corrosion for air or water ingress; full-core fuel failure and fission product release (gases and metals)			
GETTER	Fuel failure and fission metal release			
ASTEC	RN transport in reactor building			
PC-COSYMA	Off-site radiological doses			



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Comparison of FSV Predicted and Measured Kr-85m Release





Comparison of FSV Predicted and Measured Fission Metal Release



Measured and Calculated Noble Gas Release from THTR at 40% Power





Measured and Predicted Fission Gas Release from Peach Bottom Core 2





Measured and Predicted Cs Plateout Distributions in Peach Bottom HTGR



Measured and Predicted Cs Plateout Profiles in In-Pile Loop Test SAPHIR P11



- German-funded tests in CEA in-pile PEGASE loop
- Primarily a plateout test program
- Spherical fuel elements
- HEU (Th,U)O₂ BISO fuel (reference THTR fuel)
- Sorption control at high temperatures
 - ✓ Material effects
- Mass transfer control at lower temperatures
 - ✓ Flow effects
- KFA plateout predictions with PATRAS code
- Limited publication of test data



Comparison of DAMD Code Predictions with VAMPYR Plateout Data

The PBMR code DAMD (Dust and Activity Migration and Distribution) predicts RN and dust transport



Comparison between VAMPYR I V09 test and DAMD for 131I Plate-out Comparison between VAMPYR II test 1 and DAMD for 110mAg Plate-out

Both VAMPYR I (probe) and VAMPYR I I (loop) utilized AVR primary coolant as RN source





Measured and Predicted Ag-110m Plateout in COMEDIE BD-1 Heat Exchanger



Ag sorptivity of Alloy 800 over predicted (0 – 70 cm); mass transfer coefficient well predicted (>70 cm)





Measured and Predicted Liftoff in COMEDIE BD-1 Loop Test

Nuclide		Cumulative Liftoff Fraction (%)				
		SR = 0.7	SR = 1.7	SR = 2.8	SR = 5.6	
I-131	Meas.	0.077	0.10	0.11	0.13	
	Pred.	0.15	0.16	0.53	2.1	
Cs-137	Meas.	0.014	0.021	0.030	0.11	
	Pred.	0.19	0.29	0.48	1.1	
Cs-134	Meas.	0.015	0.020	0.028	0.096	
	Pred.	0.19	0.29	0.48	1.1	
Ag-110m	Meas.	0.015	0.019	0.043	0.23	
	Pred.	0.010	0.32	0.90	2.8	
Sr-90	Meas.	0.16	0.36	0.56	0.74	
	Pred.	0.54	0.56	1.2	4.2	

Shear Ratio (SR) = wall shear stress during blowdown/wall shear stress during normal operation SR <1.1 during DBDA in steam-cycle MHTGR; test data @ SR = 1.7 most relevant 5% liftoff assumed when deriving fuel performance requirements for prismatic cores



RN Transport Technology Development

- Existing RN transport knowledge base and design methods are sufficient for conceptual and preliminary designs
- Additional data needed to complete code validation
 - Single-effects data for component model upgrades
 - Independent integral data for code validation

• NGNP/AGR Fuel Plan defines requisite tests for prismatic HTGRs

- Key single-effects tests
 - In-pile irradiation tests with known failure fraction
 - Postirradiation heating tests (isothermal)
 - Laboratory sorption measurements for matrix, graphite and metals
 - Out-of-pile loop tests
- Key integral validation tests
 - In-pile irradiation tests with known failure fraction
 - Postirradiation heating tests (thermal transients)
 - In-pile loop tests

• Technology program for pebble-bed HTGRs being developed





Summary

- HTGRs employ multiple RN release barriers to meet RN control requirements and to provide Defense-in-Depth
- RN transport in HTGRs has been extensively investigated
- Design methods are available to predict RN transport from fuel kernel to site boundary
- Codes are not completely validated
- Focused technology development needed to complete code validation
- Current methods sufficient for conceptual & preliminary designs





Suggested Reading

"Fuel Performance and Fission Product Behavior in Gas Cooled Reactors," TECDOC-978, International Atomic Energy Agency, 1997

"A Review of Radionuclide Release from HTGR Cores during Normal Operation," EPRI Report 1009382, EPRI, 2004

"Plate-Out Phenomena in Direct-Cycle High Temperature Gas Reactors," EPRI Report 1003387, EPRI, 2002

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"PBMR Radionuclide Source Term Analysis Validation Based On AVR Operating Experience," <u>Proceedings HTR2008: 4th International Topical Meeting on High Temperature</u> <u>Reactor Technology, September 28 - October 1, 2008, Washington, D.C.</u>

"Technical Program Plan for the NGNP/AGR Fuel Development and Qualification Program" INL/EXT-05-00465, Rev. 2, Idaho National Laboratory, 2008

