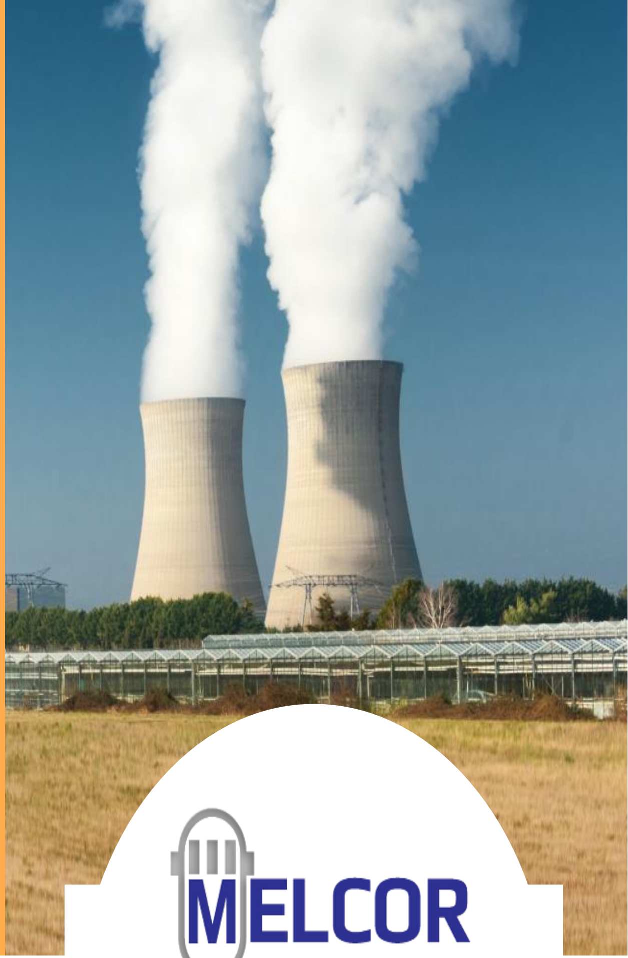
Securing the future of Nuclear Energy



Fission Product & Radionuclide Release

2024 European MELCOR Users' Group Meeting

April 15th-18th, 2024

SAND2024-03307C

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Overview

Review LWR and non-LWR fission product release

Four basic release models/options

- CORSOR, CORSOR-M, CORSOR-Booth, & Modified CORSOR-Booth
- Validation

Generalized release model

Specialized release models

- TRISO fuel diffusional release model
- Metal fuel release model
- Structural Ag-In-Cd control rod material

Reminder

User initializes fission product mass (sorted by radionuclide class) over core cells in fuel and/or gap (RN1_FPN/RN1_GAP)

User sets gap release conditions (RN1_GAP00)

User picks release model (RN1_FP00)

- Governs release from…
  - Intact fuel material in FU component,
  - Refrozen fuel material (as conglomerate)
  - Fuel material as other components (PD and MP)
  - Possibly structural materials as non-FU components (nonradioactive)

TRISO and metal fuel release models require other/different inputs in COR and may or may not use RN1_FP00 elections

CORSOR and CORSOR-M Models

CORSOR correlates fractional release rate [-/min] as:

$$\boldsymbol{R = Ae^{B*T}, \quad for\ T > T_i}$$

- A and B are empirical constants, T is component temperature [K]
- T_i is a lower temperature limit
- RN1 SC 7101 for class-wise constants and limits

CORSOR-M correlates fractional release rate [-/min] as:

$$\boldsymbol{R = k_0 e^{-Q/(R*T)}}$$

- $k_0\ [{}^{-}/_{min}]$ and $Q\ [{}^{kcal}/_{mol}]$ differ by radionuclide class
- T is component temperature [K]
- R is $1.987 * 10^{-3}\ \left[\frac{kcal}{mol*K}\right]$
- RN1 SC 7102 for class-wise constants and limits

CORSOR-Booth Model

Calculate class-wise release from scaling of mass transport limited Booth model prediction for Cesium release

- Booth model
  - Approximate Fick's law solution on a sphere of radius a
  - Use Arrhenius diffusion data from Cs (RN1 SC 7106)

$$D = D_o \exp(-Q/RT)$$

$$f = 6\sqrt{\frac{D' t}{\pi}} - 3\,D' t \qquad \text{for } D' t < 1/\pi^2$$

$$f = 1 - \frac{6}{\pi^2}\exp(-\pi^2 D' t) \qquad \text{for } D' t > 1/\pi^2$$

where
$D' t$ $= Dt/a^2$ (dimensionless)
a = equivalent sphere radius for the fuel grain

$$Release\ rate_{Cs} = \frac{[(f\sum D'\Delta t)_{t+\Delta t} - (f\sum D'\Delta t)_t]V\rho}{(1-f)\Delta t}$$

- Gas phase mass transport limitation

$$\frac{1}{M_{Cs}} = \frac{D_{fuel}RT}{A_{fuel}NuD_{k,gas}P_{k,eq}}$$

where
D_{fuel} = diameter of fuel pellet
A_{fuel} = fuel rod flow contact area
$D_{k,gas}$ = diffusivity of class k in the gas mixture
Nu = Nusselt number
$P_{k,eq}$ = equilibrium vapor pressure of class k at temperature T

- Overall:

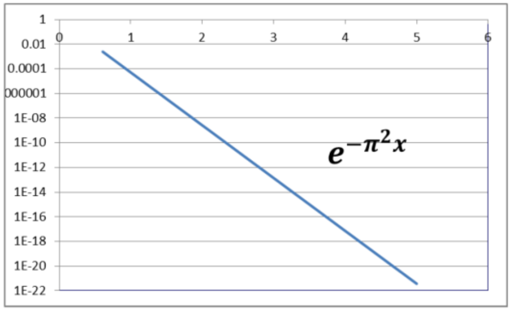
$$DIFF_{Cs} = \left[\frac{1}{\text{Release rate}_{Cs}} - \frac{1}{M_{Cs}}\right]^{-1} \qquad DIFF_k = DIFF_{Cs}S_k$$

See RN1 SC 7103 & 7107

CORSOR-Booth Model

Potential underflow issue

- Quantity $\sum D'\Delta t)$ increases monotonically
- Evaluate quantity $\exp(-\pi^2 D' t)$ to 0 for $\sum D'\Delta t) > 5$
- Effectively halt Cs (and all RN class) release



Release fraction

- MELCOR internally tracks RFRAC, the “fraction remaining” of Cs
- For a more consistent calculation, solve Booth Cs release rate as:

$$Release\ rate_{Cs} = \frac{[(f\sum D'\Delta t)_{t+\Delta t} - (f\sum D'\Delta t)_t]V\rho}{(1-f)\Delta t} \longrightarrow Release\ rate_{Cs} = \frac{V\rho}{(1-RFRAC)}\frac{d(RFRAC)}{dt}$$

- Where time derivatives of RFRAC come from $f(t)$, e.g.

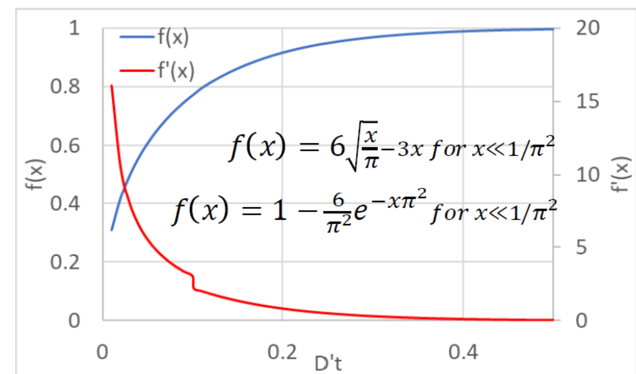
$$f = 1 - \frac{6}{\pi^2}\exp(-\pi^2 D' t) \quad \text{for } D' t > 1/\pi^2 \longrightarrow \frac{d(RFRAC)}{dt} = \pi^2 D'(1-RFRAC), \quad for\ D't > \frac{1}{\pi^2}$$

CORSOR-Booth Model

Potential issues with Booth approximation

Derivative of approximation to $f(t)$ discontinuous at $t = \frac{1}{\pi^2 D'}$

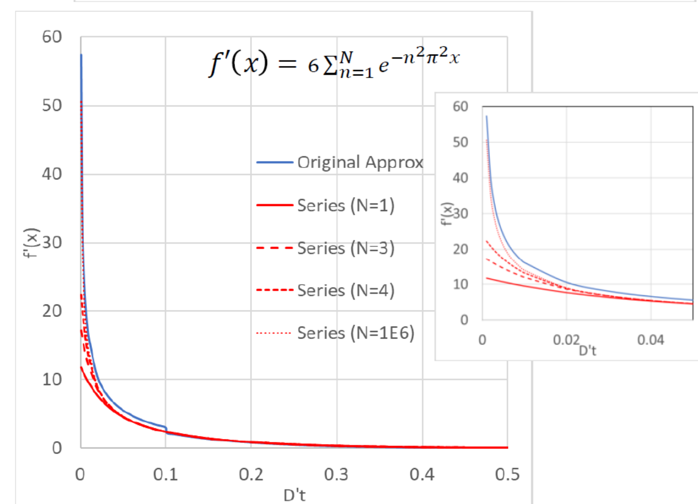
- Error ~ 50%
- Implications for fitting?
- Interpretation in calculated release?



Could use a series solution

$$f(x) = 1 - \frac{6}{\pi^2} \sum_{n=1}^{\infty} \frac{1}{n^2} e^{-n^2 \pi^2 D'}$$

- Better agreement at discontinuity (3 or 4 terms may be sufficient)
- As many as 10 terms needed for good agreement as $D't \rightarrow 0$



CORSOR-Booth Model

Booth scaling factors not derived for use as multipliers to the Cs release rates, thus a new Booth model was developed

New ORNL-Booth Model

- Diffusion coefficients scaled to Cs diffusion coefficients and independent diffusion calculation for each class

$$= \int_0^{D't} \frac{f_k'}{1 - f_k} \cdot (1 - f_k)$$

Existing modified ORNL-Booth Model

- Scale RN release rates to Cs Booth diffusion release rate

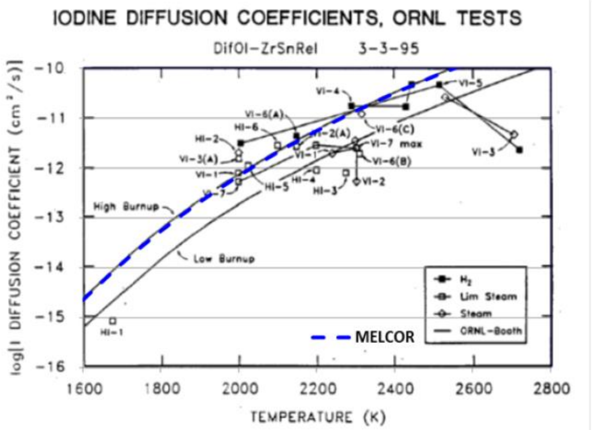
$$= \int_0^{D't} S_k \frac{f_{Cs}'}{1 - f_{Cs}} \cdot (1 - f_k)$$

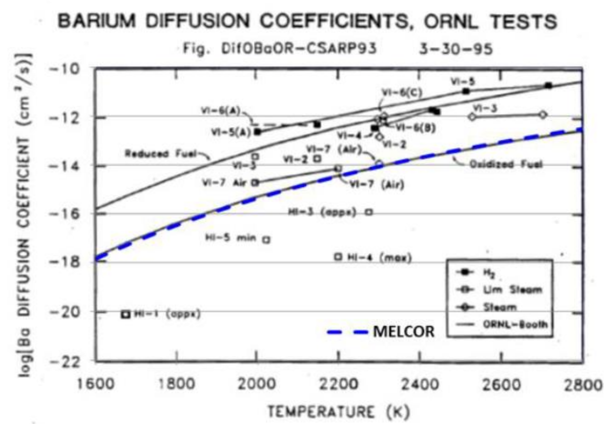
CORSOR-Booth Model

Corrected Booth modeling with scaled diffusion coefficients

- MELCOR diffusion coefficients scaled with ORNL Data
  - $D_{class} = S_{class} D_0 \, e^{-\frac{Q}{RT}}$
- Perform diffusion calculation for each class using appropriate diffusion coefficient
  - ICRLSE=7,-7

Diffusion Coeff, D_0	1×10^{-6} m^2/sec
Activation Energy, Q	3.814×10^5 Joule/mole
S_{XE}	1
S_{Cs}	1
S_{Ba}	4×10^{-4}
S_I	0.64
S_{Te}	0.64
S_{Ru}	0.0025
S_{Mo}	0.2
S_{Ce}	4×10^{-8}
S_{La}	4×10^{-8}
S_{Cd}	.25
S_{Sn}	.16





CORSOR-Booth Model

Corrected Booth modeling – validation

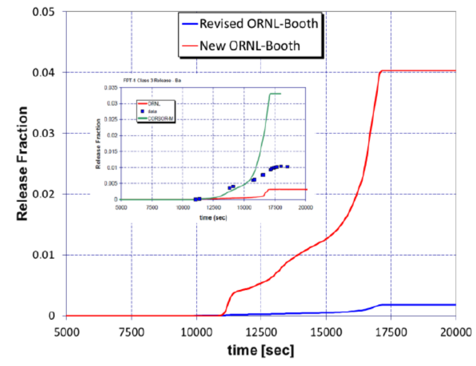
- Validation of model against FPT-1 experiment
  - ‘New ORNL-Booth’ (ICRLSE=-7,7) represents modeling based on scaling diffusion coefficients
  - Release is only changed for those RNs with diffusion scale factors much less than unity
    - Comparison of Barium release
      - Diffusion coefficient is orders of magnitude smaller than Cs (scale factor = $4x10^{-4}$)
      - Predicts larger release fraction than observed
        - » May need to be adjusted
      - Predicts much larger release fraction than ‘Revised ORNL-Booth’ (ICRLSE=-5,5) though relative error is about the same
      - Note inconsistencies in ‘Total’ and ‘Total Released’ in text output
        - » Round-off issues leads to 30% error
        - » May account for error in release fraction (in part)

RELEASED RADIOACTIVE RADIONUCLIDE MASS AUDIT - MASSES IN KG

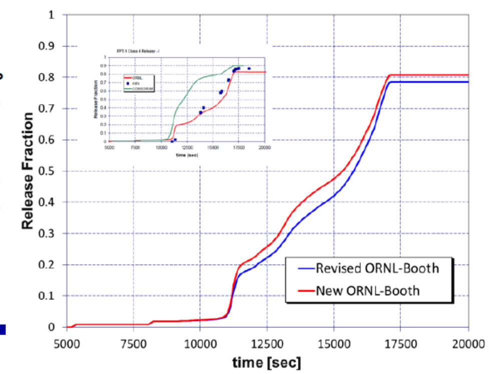
CLASS	TOTAL	TOTAL RELEASED	AEROSOL MASSES GAS	AEROSOL MASSES LIQUID	VAPOR MASSES GAS	VAPOR MASSES LIQUID	DEP MASS	FILTERS	CHEM AB	PHYSDEP
XE	2.819E-02	2.819E-02	0.000E+00	0.000E+00	2.819E-02	0.000E+00	0.000E+00	0.000E+00	0.000E+00	0.000E+00
CS	9.708E-03	1.181E-02	0.000E+00	4.256E-03	2.224E-20	0.000E+00	5.242E-03	0.000E+00	2.095E-04	0.000E+00
BA	4.704E-04	6.211E-04	0.000E+00	1.826E-04	0.000E+00	0.000E+00	2.878E-04	0.000E+00	0.000E+00	0.000E+00

- Comparison of I_2 Release
  - Very Slight differences
  - Diffusion coefficient scale factor close to unity (0.65)

Barium



Iodine



CORSOR-Booth Model

Corrected Booth modeling – gaseous mass transport

$$\dot{m}_k = \left[Nu \frac{D_{k,gas}}{D_{fuel}} \right] \cdot A_{fuel} \cdot \left(\frac{P_{k,eq}}{RT} - \cancelto{0.0}{\rho_{k,free}} \right)$$

Gas-phase mass transfer from heat transfer analogy (free stream density of all RNs assumed zero)

- CORSOR-Booth Model (ICRLSE=3,-3)
  - Mass transfer term removed from effective release rate before scaling other RNs

$$ReleaseRate_{Cs} = \frac{[f(\Sigma_{t+\Delta t} D' \Delta t) - f(\Sigma_t D' \Delta t)]}{(1 - RFRAC)\Delta t} V\rho$$

$$DIFF_{Cs} = \left[\frac{1}{ReleaseRate_{Cs}} - \frac{1}{\dot{m}_{Cs}} \right]^{-1}$$

$$DIFF_k = DIFF_{CS} \cdot S_k$$

- Revised CORSOR-Booth Model (ICRLSE=5,-5)
  - Mass transfer term not removed from Cs release rate
    - Assumes ReleaseRate only accounts for diffusion

$$DIFF_k = ReleaseRate_{CS} \cdot S_k$$

- Mass transfer term added back to each RN $\quad \dot{m}_k = [1/DIFF_k + 1/\dot{m}_k]^{-1}$

CORSOR-Booth Model

Gaseous mass transport relative to UO_2 in volume

- Note that RM indicates Cs release rate is fractional release multiplied by ρV
  - Where V is volume and ρ is UO_2 molar density
  - New model uses actual class inventory
- Note however that release rate is divided by ρV before multiplying by RN inventory to obtain mass released
- Even so, scaling by ρV is important in comparing diffusion release rate with gas release rate

Old Modeling

$$ReleaseRate_{Cs} = \frac{[f(\sum_{t+\Delta t} D'\Delta t) - f(\sum_t D'\Delta t)]}{(1 - RFRAC_{Cs})\Delta t}\rho V$$

$$\dot{f}_k \text{ (fraction/s)} = \frac{\dot{m}_{tot,k}}{\rho V}\left[F - \frac{P_{k,bulk}}{P_{k,eq}}\right]$$

$$\dot{m}_k = [1/DIFF_k + 1/\dot{m}_k]^{-1}$$

New Modeling

$$ReleaseRate_{Cs} = \frac{[f(\sum_{t+\Delta t} D'\Delta t) - f(\sum_t D'\Delta t)]}{(1 - RFRAC_k)\Delta t}Mass_{k,component}$$

$$\dot{f}_k(fraction/s) = \frac{\dot{m}_{tot,k}}{Mass_k}\left[F - \frac{P_{k,bulk}}{P_{k,eq}}\right]$$

$$\dot{m}_k = [1/DIFF_k + 1/\dot{m}_k]^{-1}$$

CORSOR-Booth Model

Strictly valid in intact fuel as release depends on history

- Release fraction for intact fuel well characterized
- No moving material associated with intact fuel
- All other components bearing fuel material only do so after a transport occurs (conglomerate or PD)
- MELCOR blends release fractions after transport (meaningful?)

Unoxidized Zr can lead to reduction in release rate of Te

- Te reacts with Zr to form low vapor pressure products
- Te release rate reduced by a multiplier until mass of unoxidized intact metal CL (Zr) falls below a cut-off fraction of total intact CL
- Parameters are SC7105 and SC7107

T. Nakamura and R. A. Lorenz, "Effective Diffusion Coefficients Calculated from ORNL FP Release Test Results," Oak Ridge National Laboratory Research Paper (April 1989).

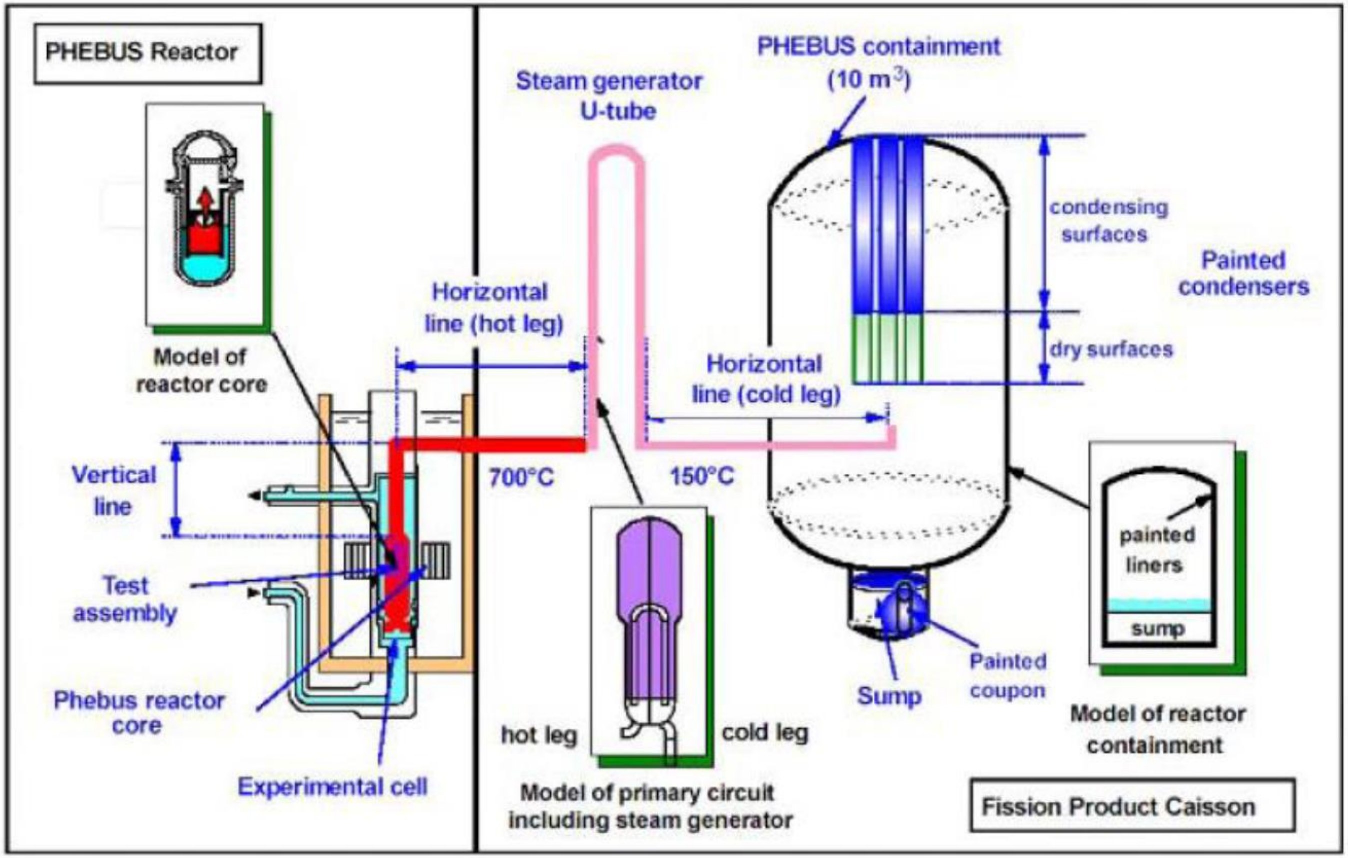
CORSOR-Booth Model

Original CORSOR-Booth class scaling factors were modified based on ORNL-VI results

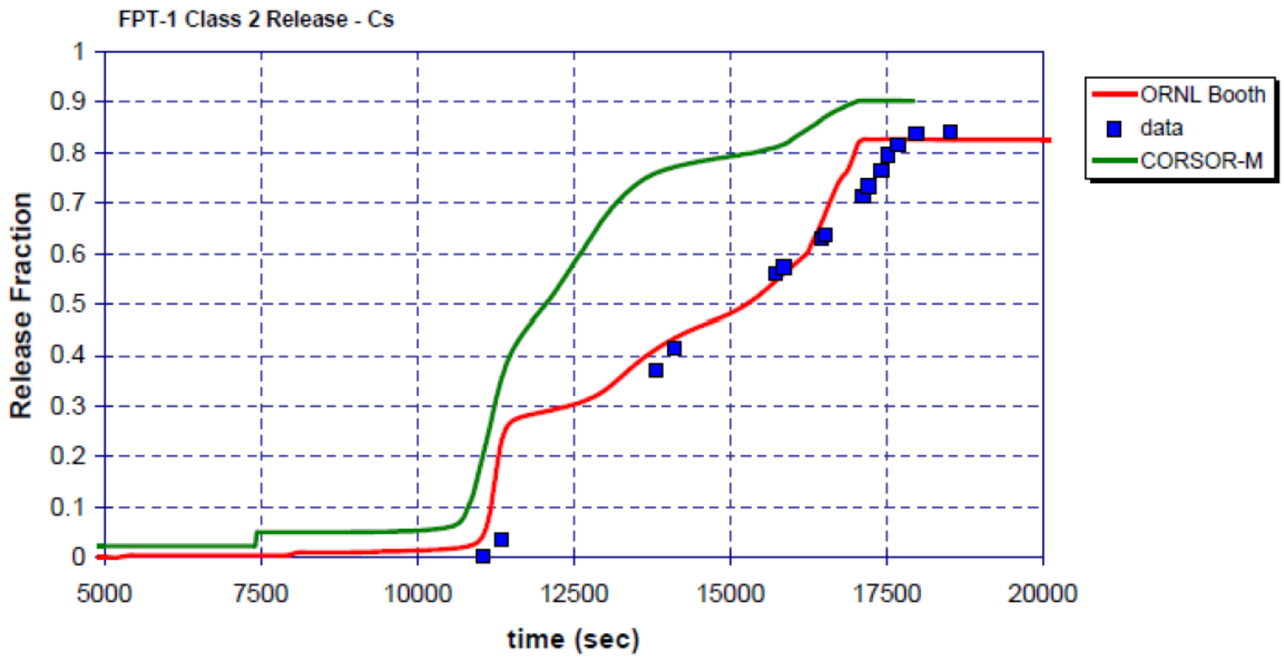
- Dominant Cs species changed from CsI to CsMoO4
- FP releases adjusted based on Phebus-FPT1
- Results compared again to ORNL-VI and VERCORS (2 and 4)

Modified CORSOR-Booth shows improvement over original CORSOR-Booth generally

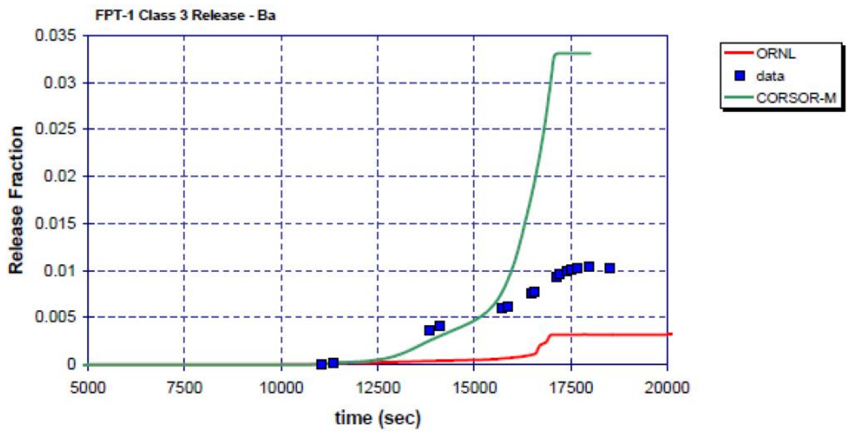
Phebus Test Facility

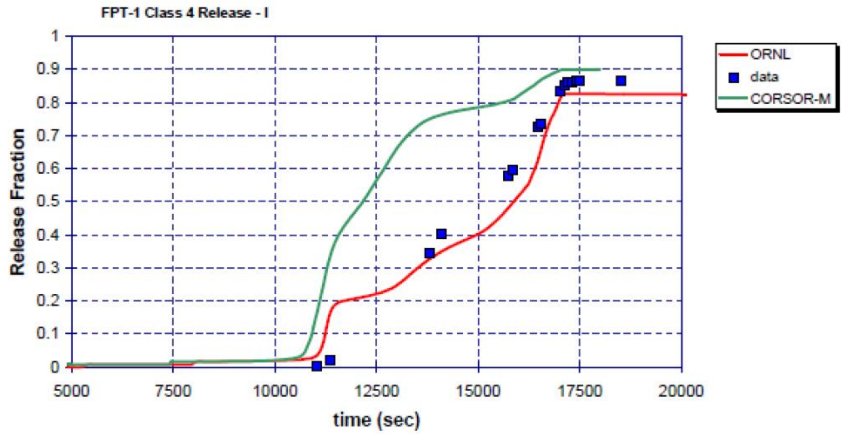


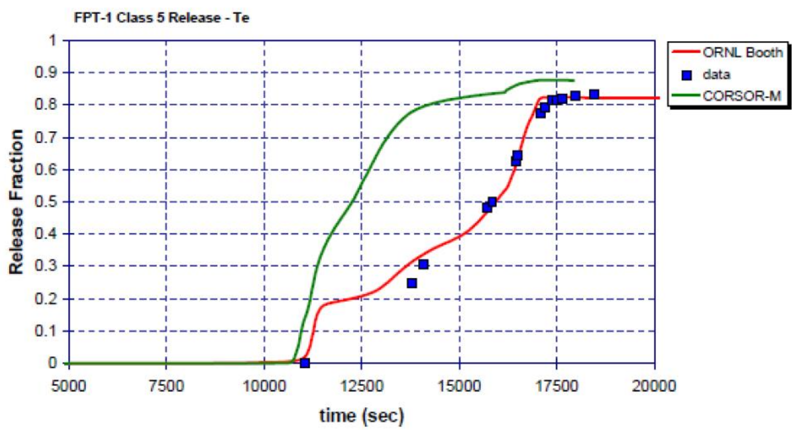
Phebus-FPT1 Cs

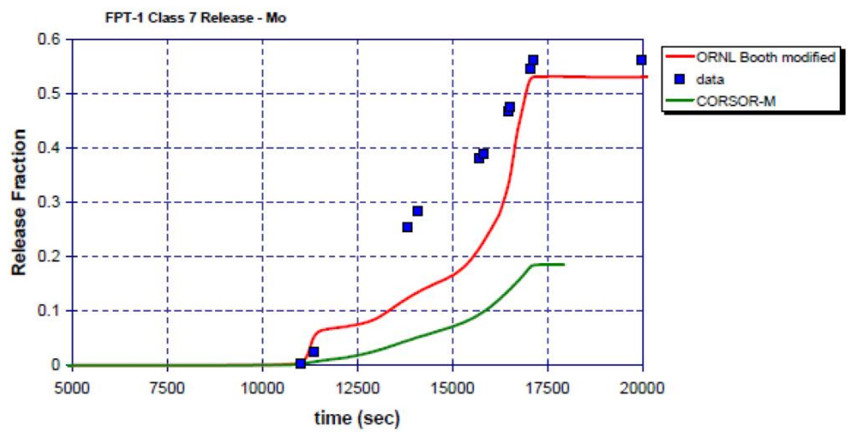


Phebus-FPT1 Other Classes



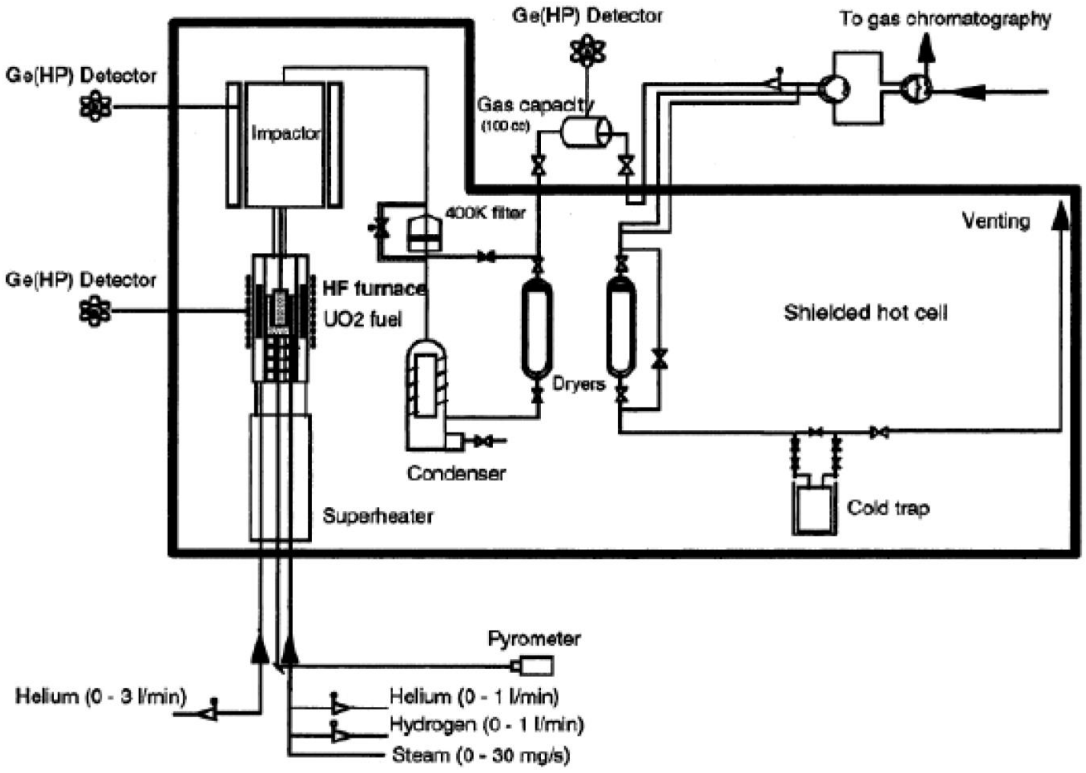




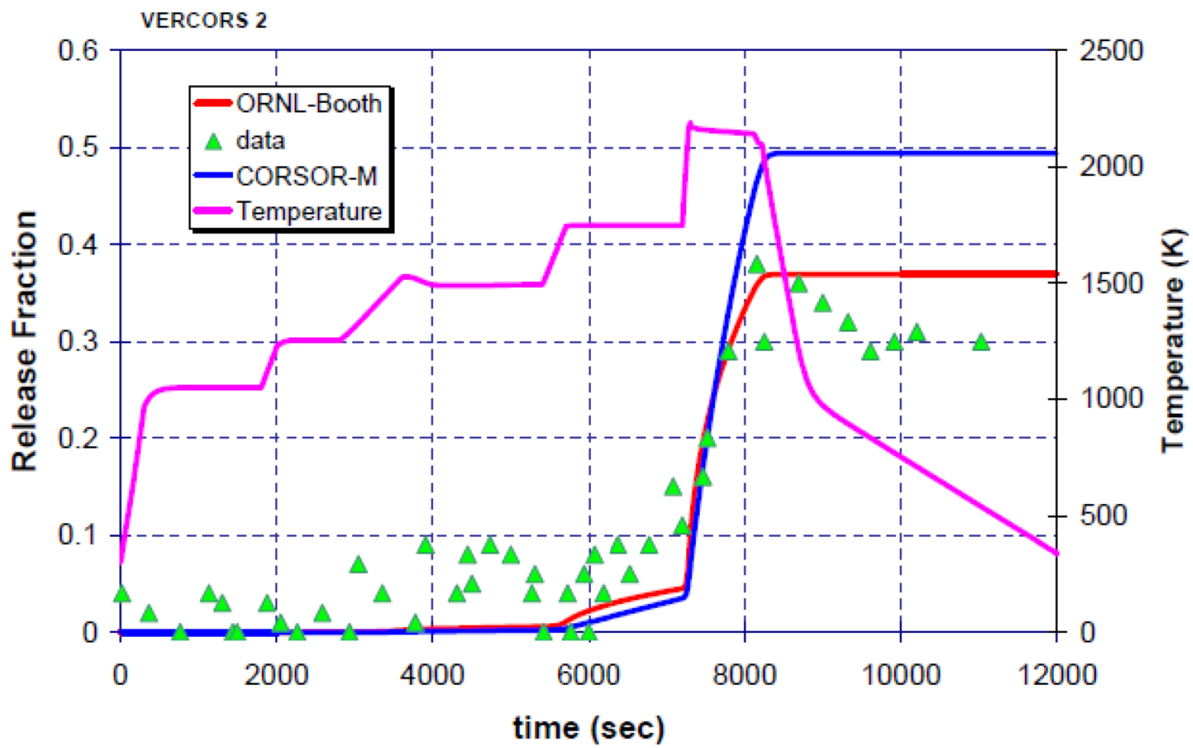


VERCORS Test Facility

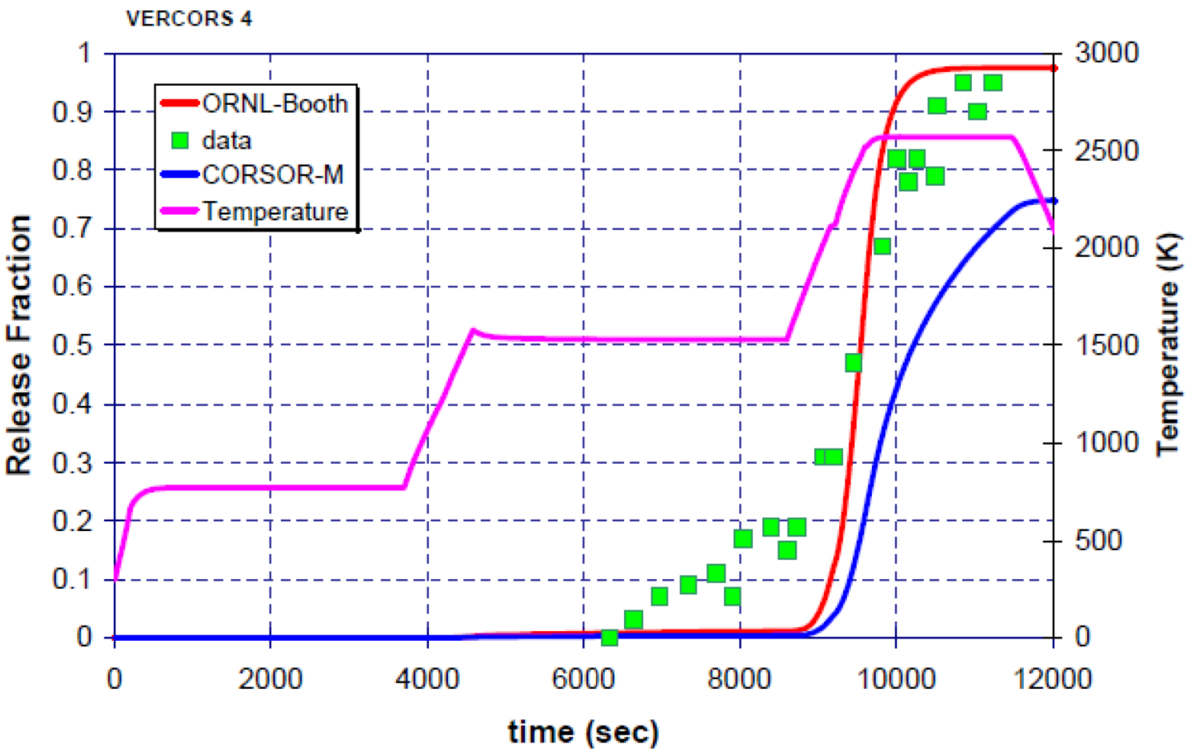
G. Ducros et al. / Nuclear Engineering and Design 208 (2001) 191–203



VERCORS-2 Cs


VERCORS 2
ORNL-Booth
data
CORSOR-M
Temperature
Release Fraction
Temperature (K)
time (sec)
0
0.1
0.2
0.3
0.4
0.5
0.6
0
500
1000
1500
2000
2500
0
2000
4000
6000
8000
10000
12000

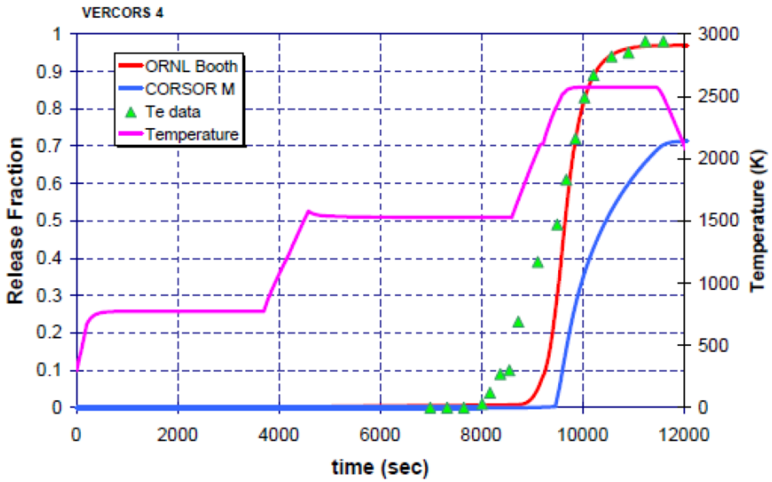
VERCORS-4 Cs



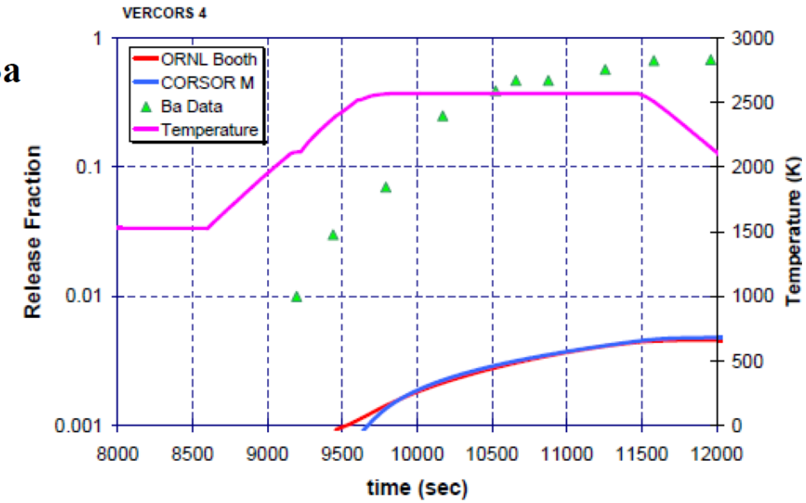
VERCORS 4
ORNL-Booth
data
CORSOR-M
Temperature
Release Fraction
Temperature (K)
time (sec)
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
1
500
1000
1500
2000
2500
3000
2000
4000
6000
8000
10000
12000

VERCORS-4 Other Classes

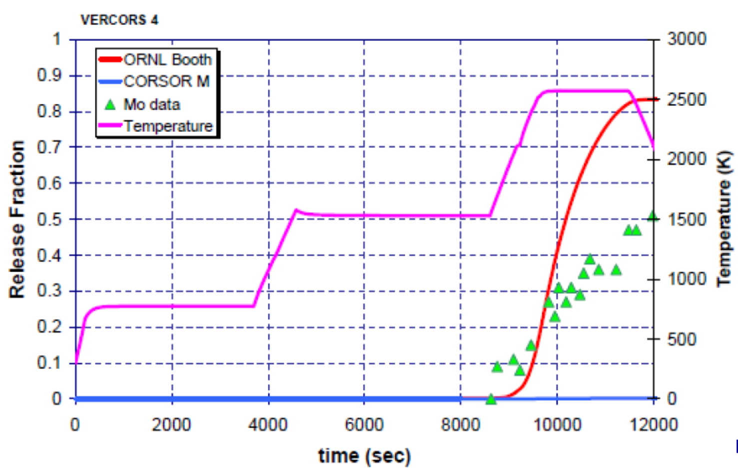
Te



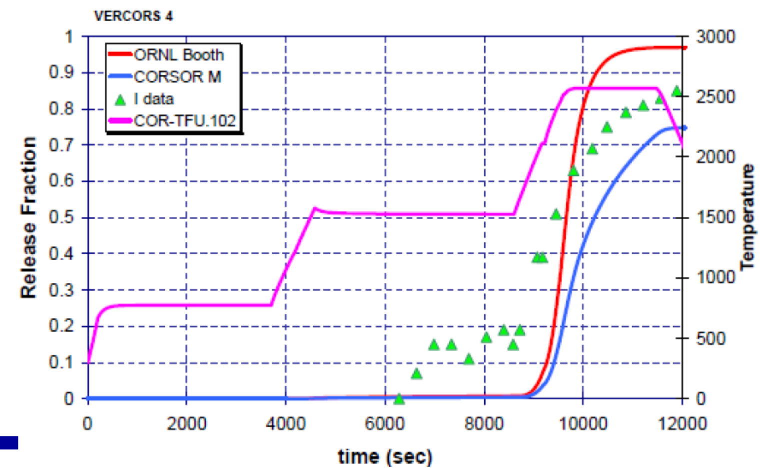
Ba



Mo



I



Generalized Release Model

An alternative offering flexibility and customizability

- A "burst" component, cumulative burst release fraction:

$$FB_{j,i} = a\_burst_j\left(c_0 + c_1 * T_i + c_2 * T_i^2 + c_3 * T_i^3\right)$$

Where

T_i is the fuel temperature that existed during the time interval Dt_i

c_0, c_1, c_2, c_3 are constant coefficients provided in user input

a_j is a constant class dependent coefficient provided in user input.

- A "diffusion" component, cumulative diffusive release fraction:

$$FD_{j,i} = b\_diff_j\left(FD_{j,i-1} + \left(1 - FB_{j,i-1} - FD_{j,i-1}\right) \cdot \left[1 - e^{-kd_{j,i} \cdot \Delta t_i}\right]\right)$$

Where

$FD_{j,i}$ is the cumulative fraction of diffusive fission product released up to time t_i

B_diff_j is a constant class dependent coefficient provided in user input

$FD_{j,i-1}$ is the cumulative fraction of diffusive fission product released up to time t_{i-1}

FB_i is the cumulative fraction of burst fission product released up to time t_i

$\left[1 - e^{-kd_{j,i} \cdot \Delta t_i}\right]$ is the fractional release due to diffusion during the time interval Dt_i

$kd_{j,i}$ is the release rate coefficient for fission product class j calculated using the temperature, Ti, that existed during the time interval Dt_i

$$kd_{j,i} = A_j e^{-B_j/(RT_i)}$$

Where A_j and B_j are class dependent coefficients provided in user input.

Generalized Release Model

Total cumulative fission product release fraction at t_i for j is:

$$F_{j,i} = d\_total_j \cdot (FB_{j,i} + FD_{j,i})$$

User supplied class-dependent multiplier

Cumulative release cannot exceed amount available

$$FB_{j,i} = FB_{j,i-1} \ and \ FD_{j,i} = FD_{j,i-1} when \ FD_{j,i} \geq 1.0$$

Time derivative of cumulative burst release ≥ 0, and if temperature decreases, cumulative burst remains constant

$$FB_{j,i} = FB_{j,i-1} \ when \ T_{i-} \geq T_{B-max} \ or \ T_{melting}$$

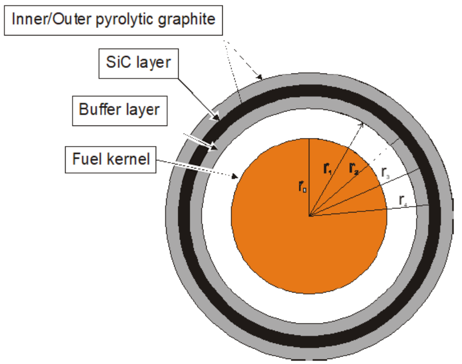
Cumulative burst release reaches maximum when temperature reaches the lower of T_{B-max} or $T_{melting}$

$$FB_{j,i} = FB_{j,i} \ when \ T_i \geq T_{B-max} \ or T_{melting}$$

TRISO Fuel Release

HTGR (PBR/FHR and PMR) fuel element release entails phenomena not occurring in LWRs

- Failure and release spread out over time
  - Low level operational release (circulating activity)
  - Longer accident release time scale
- Graphite dust present in primary



TRISO diffusional fission product release model considers

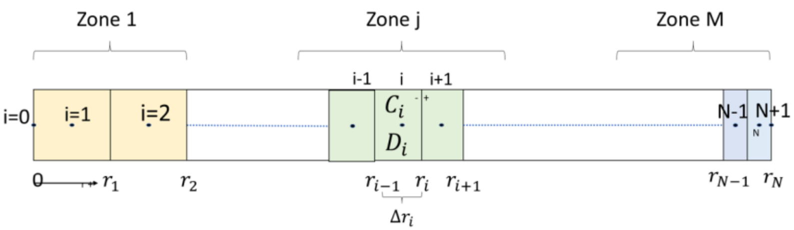
- Diffusion from kernel through to fuel element matrix and out
- Fission product recoil
- Tramp Uranium (contamination)
- TRISO particle failure (burst and subsequent diffusion)
- Time and temperature history of diffusion
- Graphite dust generation and transport (e.g. as aerosol)

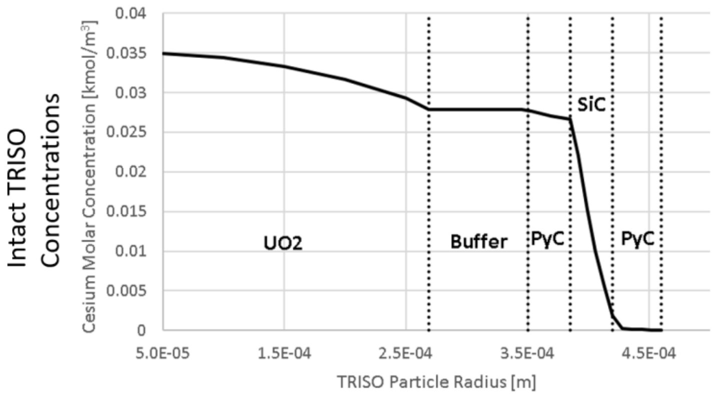
TRISO Fuel Release

Intact TRISO Particles

- 1-D FV diffusion solver, multiple zones (materials)
- $D(T)$ in Arrhenius form

$$\frac{\partial C}{\partial t} = \frac{1}{r^n}\frac{\partial}{\partial r}\left(r^n \boldsymbol{D}\frac{\partial C}{\partial r}\right) - \lambda C + \beta \qquad D(T) = D_0 e^{-\frac{Q}{RT}}$$





Diffusivity Data Availability

Radionuclide	UO_2	UCO	PyC	Porous Carbon	SiC	Matrix Graphite	TRISO Overall
Ag	Some	Not investigated	Some	Not found	Extensive	Some	Extensive
Cs	Some		Some		Extensive	Some	Some
I	Some		Some		Some	Not found	Not found
Kr	Some		Some		Not found	Some	Some
Sr	Some		Some		Extensive	Some	Some
Xe	Some		Some		Some	Some	Not found

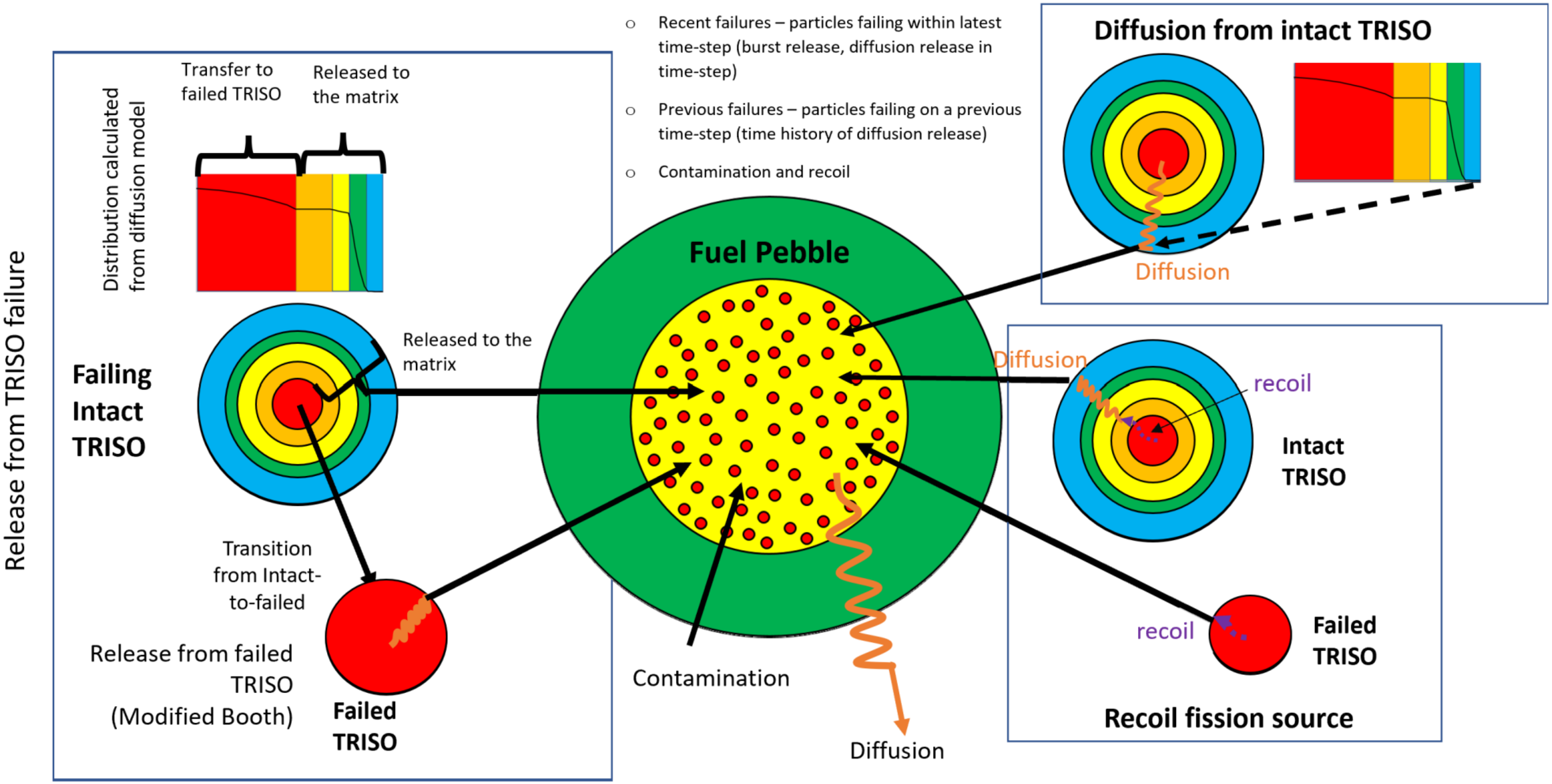
Data used in the demo calculation
[IAEA TECDOC-0978]

	FP Species							
	Kr		Cs		Sr		Ag	
Layer	D (m²/s)	Q (J/mole)	D (m²/s)	Q (J/mole)	D (m²/s)	Q (J/mole)	D (m2/s)	Q (J/mole)
Kernel (normal)	1.3E-12	126000.0	5.6-8	209000.0	2.2E-3	488000.0	6.75E-9	165000.0
Buffer	1.0E-8	0.0	1.0E-8	0.0	1.0E-8	0.0	1.0E-8	0.0
PyC	2.9E-8	291000.0	6.3E-8	222000.0	2.3E-6	197000.0	5.3E-9	154000.0
SiC	3.7E+1	657000.0	7.2E-14	125000.0	1.25E-9	205000.0	3.6E-9	215000.0
Matrix Carbon	6.0E-6	0.0	3.6E-4	189000.0	1.0E-2	303000.0	1.6E00	258000.0
Str. Carbon	6.0E-6	0.0	1.7E-6	149000.0	1.7E-2	268000.0	1.6E00	258000.0

Iodine assumed to behave like Kr
CORSOR-Booth LWR scaling used to estimate other radionuclides

TRISO Fuel Release

- Recent failures – particles failing within latest time-step (burst release, diffusion release in time-step)
- Previous failures – particles failing on a previous time-step (time history of diffusion release)
- Contamination and recoil



Metal Fuel Release

Begin: as-fabricated pin is clean/unburned/unswollen

Pre-transient: burned pin transforms as-fabricated pin

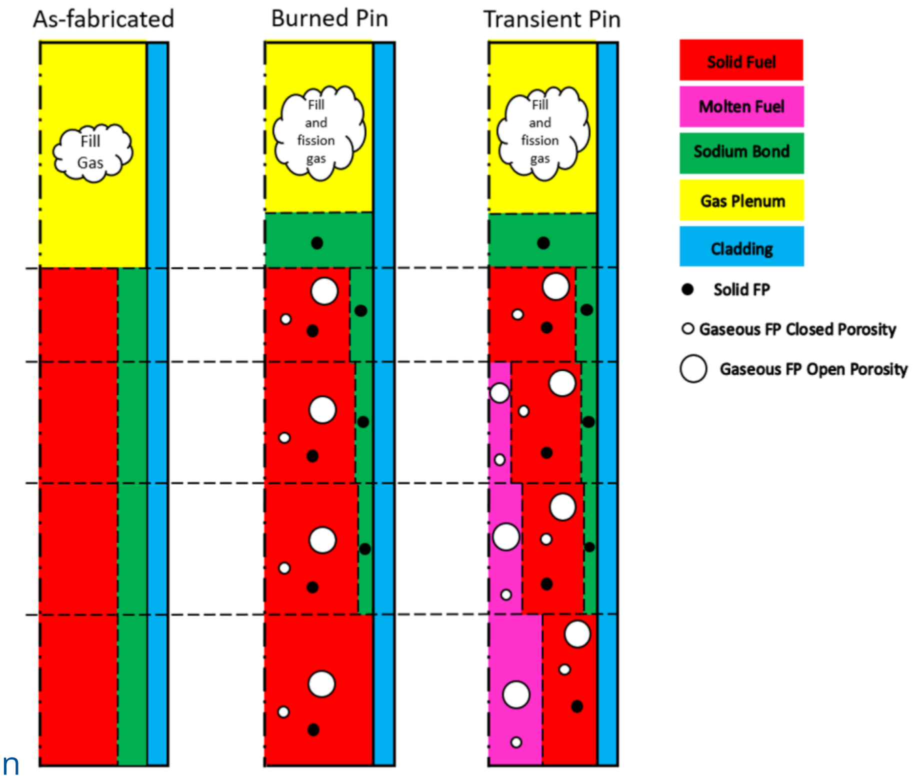
- Bond sodium contents and thickness/volume
- Pin gas plenum contents and volume
- Fuel porosity distribution, contents, volume
- One-time computation of redistributions

Transient: compute in-pin dynamics

- Fuel can melt, create cavities – possibly contiguous
- Fission gas (closed to open, solid fuel to molten)
- Fuel can release fission products to gap, plenum

Accident: pin failure then severe core degradation

- CL gap release, CL/CN candling, eventual collapse
- FU/CL collapse, PD/MP formation, subsequent relocation



Ag-In-Cd Control Rod Release

FPT-1 post-test analysis demonstrated the importance of predicting structural aerosol releases from CR materials

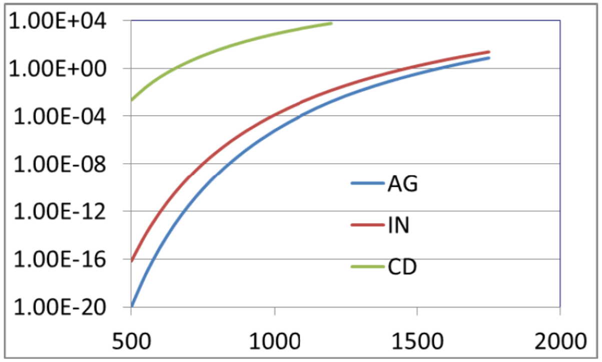
MELCOR has 1 material (e.g. primary of NS component used for control rods) representing all three of Ag-In-Cd

- Material vapor pressure taken as the lowest of Ag, In, or Cd
- Material composition does not change when in reality Cd, In, and Ag would vaporize, respectively, and composition would change
- Vaporization model therefore entails some simplification

Mass diffusion in bulk gas controls vaporization

- No NS component cooling by vaporization
- No vapor condensation allowed

$$\dot{m}_v = h_m A_c M_{Ag}\left(\frac{P_v(T_c)}{RT_c} - \frac{P_{Ag}(T_b)}{RT_b}\right)$$



Ag-In-Cd Control Rod Release

COR_CR record to enable

COR_CR ACTC

User must specify 3 new RN classes (AG-CR, IN-CR, CD-CR)

- DCH_CL and DCH_EL
- Vapor pressure (SC7110)
- Diffusion (SC7111)
- Mol. Weights (SC7120)

(1) IAICON

Turns on the silver release model

(a) 0 or NACT
Model is not active. No additional fields are required.

(b) 1 or ACTC
Model is active, vaporization is allowed from candling material only

(c) 2 or ACTDC
Model is active, vaporization is allowed from both candling material and conglomerate

(type = integer / character*5, default = 0, units = none)

The following fields are required if and only if IAICON = ACTC or 1, ACTDC or 2. If they are not set, then the default values will be used:

(2) ARATIO

The area ratio of break area to control rod internal cross-sectional area. This cannot be greater than 1.

(type = real, default = 0.1, units = none)

(3) AKFRCT

The flow loss coefficient used in the release velocity calculation. If input, this will override the value calculated according to the formula given in the reference manual.

(type = real, default = 0.32*(1.0 - ARATIO), units = none)

Summary

Reviewed CORSOR, CORSOR-M, CORSOR-Booth, and modified CORSOR-Booth models

Reviewed the generalized release model

Reviewed aspects of certain non-LWR specific release models

- TRISO fuel and fuel elements (diffusional fission product release)
- Metallic fuel as in SFRs

Reviewed structural release model for Ag-In-Cd in CRs

NOT mentioned…fission product release in CAV (VANESA)