

Iodine spiking model for pressurized water reactors

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Received 28 February 1996; accepted 13 November 1996

Abstract

An analytical treatment has been developed to describe the phenomenon of iodine spiking in the reactor coolant system during reactor shutdown with defective PWR fuel rods. The iodine mass inventory is conserved in the model. The mass transport of iodine in the fuel-to-clad gap is based on a diffusion mechanism, and a bulk-convective process during pressure and temperature transients. Iodine release data, obtained from the operational experience of PWR plants, have been used for model validation. The model has been employed to evaluate the methodology of the standard review plan for an accident-initiated iodine spike for a steam generator tube rupture.

1. Introduction

When a nuclear reactor fuel rod defects, the clad no longer provides a barrier between the internal rod atmosphere and the primary coolant. A leak path then exists so that coolant can enter the rod and fission products can escape into the reactor coolant system (RCS). A number of investigations have provided a better understanding of the physical processes of activity release while the reactor is operating at steady-state power [1–11]. Only a small fraction of the fission-product iodine in a defective rod is released into the RCS during constant, full-power, operation. Most of the iodine available for release is present as a liquid-water soluble deposit on the UO₂ fuel surface or inner surface of the cladding. If the temperature in the pellet-to-clad gap drops below that of coolant saturation, as during reactor shutdown, the water that has entered the rod remains in the liquid phase and leaches these deposits. The dissolved iodine can then migrate along the water-filled gap to the defect site, resulting in an increased release to the RCS. This accelerated release leads to the so-called ‘iodine spike’ [12–19]. An activity contribution to the

spike can also result from coolant depressurization and temperature transients during shutdown.

The iodine-spiking phenomenon is an important consideration in safety analysis [18,20]. For instance, in a pressurized water reactor (PWR), the high pressure coolant is circulated through heat exchanger tubes in the steam generators that represent a large fraction of the RCS boundary. The rupture of a tube will result in a reactor trip, with an enhanced release of iodine into the coolant and a direct path for release to the environment [18]. A steam generator tube rupture (SGTR) accident has in fact been designated as a design basis accident for PWRs [21].

This paper describes the development of a physical model that may be used for those type of events in which activity release due to the iodine-spiking phenomenon can occur. Previous treatments have generally ignored the time-dependent behavior for the rate of release of iodine from the defective fuel rod into the coolant [18,20]. In another approach, the release rate was modelled as a simple impulse function where mass conservation was ignored; i.e., the escape rate constant for steady-state operation was simply multiplied by a ‘spiking factor’ that was proportional to the fractional change in power (or pressure) [14]. As conceded in this latter study, a more realistic time–response function was needed because the spike was generally predicted too early. In contrast, the

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present work accounts for enhanced-diffusional release during reactor shutdown, and includes any forced-convective release that may result from temperature and pressure transients that are associated with the shutdown event. The model has been benchmarked against a database of reactor trips from PWR operational experience [12,14,20,22].

2. Model development

2.1. Steady-state release

Volatile fission products that are generated in the UO_2 fuel matrix are partially released into the fuel-to-clad gap during steady-state reactor operation. In defective fuel rods, these products migrate through the steam-filled gap toward the defect site, and eventually into the primary coolant. The release rate from the defected fuel rod to the coolant can be described with a first-order kinetic model for gap transport [1–6,8,10,11]:

$$R_c = \left(\frac{\nu}{\lambda + \nu} \right) R_f, \quad (1)$$

where R_c = release rate from the gap into the coolant (atom/s), R_f = release rate from the fuel into the gap (atom/s), ν = gap escape rate constant (s^{-1}), and λ = radioactive decay constant (s^{-1}). For defective PWR fuel rods, R_f is due predominantly to a diffusion mechanism [6,11]. There is also a small contribution from fission recoil. The escape rate constant ν depends on the defect location and size, and accounts for holdup due to physical transport along the gap to the defect site [8]. Moreover, as demonstrated in a number of in-reactor studies, this parameter also accounts for chemical holdup effects in the gap, i.e., it is particularly sensitive to the defect characteristics, owing to the relative quantity of water-to-steam around the defect site, resulting in a localized release of iodine [3,10].

During steady-state conditions, the following mass balance for the fission product inventory, N_c (atoms) in the RCS applies:

$$\frac{dN_c}{dt} = R_c - \lambda N_c - LN_c = 0, \quad (2)$$

where the rate of release into the coolant (R_c) is balanced by various losses. These losses include radioactive decay (the term containing λ), as well as coolant purification, leakage, and fission-product deposition as described by an overall loss-rate constant

$$L = \frac{F\epsilon}{M} + \frac{L_r}{M} + \alpha, \quad (3)$$

where F = cleanup system flow rate (kg/s), ϵ = efficiency of cleanup system (for iodine), M = mass of water in primary system (kg), L_r = leak rate from primary system (kg/s), and α = deposition rate constant (for iodine) (s^{-1}). Typically, the rate constants for coolant leakage and

deposition are much smaller than that for coolant purification. Using Eqs. (1) and (2), the total steady-state coolant activity resulting from x defective fuel rods is given by:

$$\lambda N_c = x \left(\frac{\lambda}{\lambda + L} \right) \left(\frac{\nu}{\lambda + \nu} \right) R_f, \quad (4)$$

where the parameters R_f and ν correspond to an 'average' defective rod.

The gap transport model is based on first-order kinetics. Here the release rate into the coolant is proportional to the gap inventory N_g [1,3–6,8,11]:

$$R_c = \nu N_g. \quad (5)$$

Using Eqs. (1) and (5), the total gap activity for the x fuel rods is:

$$\lambda N_g = x \left(\frac{\lambda}{\lambda + \nu} \right) R_f. \quad (6)$$

Eq. (6) can be used to calculate the iodine activity in the fuel-to-clad gap that is available for release during reactor shutdown, if release due to fuel cracking is negligible. Sweep gas experiments with fuel rods operating at a relatively high linear power of 55 kW/m have shown that the gap inventory is only increased by $\sim 15\%$ as a result of fuel cracking effects on shutdown for the long-lived isotope ^{133}Xe [23]. This process will therefore be negligible for PWR rods at the much lower average linear heat rating of 22 kW/m. Using Eqs. (4) and (6), the ratio of the gap activity to coolant activity is independent of the number of defective rods, i.e.,

$$\frac{\lambda N_g}{\lambda N_c} = \frac{\lambda + L}{\nu}. \quad (7)$$

Eq. (7) physically indicates that a smaller defect size (and value of ν) will result in a lower coolant activity, but a greater stored gap activity. Given the measured steady-state coolant activity, the gap activity can be predicted without knowledge of the power and burnup of the defective rod.

2.2. Reactor shutdown release

2.2.1. Diffusion and first-order kinetic models

Only a small fraction of the fission-product iodine in a defective rod is released into the coolant while the reactor is operating at constant power (Section 2.1). During reactor shutdown, the coolant which has entered the rod remains in the liquid phase and dissolves the iodine that is deposited on the internal rod surfaces. In the absence of any temperature or pressure fluctuations, this iodine leaching process can be described by either a diffusion (Appendix A) or first-order kinetic (Appendix B) process. In both representations, the release rate R (atom/s) from the defective rod into the RCS is given by the time-dependent relation [4,15,19]:

$$R(t) = k N_{go} \exp\{-(\lambda + k)t\}. \quad (8)$$

where N_{go} = initial iodine inventory in the fuel-to-clad gap (atom) and k = escape rate constant in the fuel-to-clad gap (s^{-1}). The parameter k is again dependent on the transport path length (i.e., defect location) (see Appendix A), and accounts for the delay due to the physical migration of iodine along the gap. Included in this process is any chemical trapping that may arise due to iodine reaction in the gap with the clad, fuel or other fission products. In addition, this parameter depends on the size of the gap, i.e., on whether there is an open gap present or, in contrast, if a tortuous migration path exists due to gap closure in high-burnup fuel (e.g., third cycle or higher) and/or oxidized fuel. With a large open gap, localized naturally-convective currents can arise resulting in an enhanced diffusivity (and value of k) (see Appendix A) [15].

In this representation, it is implicitly assumed that the amount of inventory in the gap (N_{go}) is fixed at shutdown and that this inventory can only be depleted. This assumption is supported by the fact that fuel cracking is an inefficient release process during the shutdown event (see Section 2.1). In addition, any thermal diffusion will be negligible as the fuel temperature is rapidly reduced on shutdown (i.e., in which the coolant temperature remains below the saturation point). Thus, the present model is only applicable for normal and off-normal situations where these specific conditions pertain (see Sections 3 and 4).

2.2.2. Forced-convection model

Iodine spiking can also result from coolant depressurization and temperature transients. When the RCS pressure is reduced or the RCS temperature is increased, non-condensable gases that are trapped in the plenum at the top of the rod can expand, thereby forcing iodine-rich water out of the rod and into the coolant [14]. The non-condensable gases in the plenum include the stable and radioactive isotopes of xenon and krypton, which are generated in the fission process, and hydrogen that is produced by coolant radiolysis and oxidation of the urania and Zircaloy cladding materials [10,24]. On the other hand, if a defect is located at the top of the rod, the gases can escape from the plenum. The rod will then entirely fill with water as the steam condenses on shutdown. With a temperature or pressure change in the RCS, the fluid density in the gap will also change, resulting in the possible expulsion of iodine-rich water.

As a consequence of gas expansion in the plenum, or water expansion in the rod, a forced-convective release will result until a pressure or temperature equalization is achieved (see Appendix C). The release rate expression for this transport process is given by (see Appendix D):

$$R(t) = k_o N_{go} \exp\{-(\lambda + f^{-1}k_o)t\}, \quad (9)$$

where

$$k_o = \frac{\Delta P(0)h^2}{12\mu l^2}, \quad (10)$$

and h = fuel-to-clad gap thickness (m), l = fuel-stack length (m), μ = fluid viscosity in the fuel-to-clad gap ($kg/m \cdot s$), and $\Delta P(0)$ = pressure differential between the coolant and internal rod atmosphere at the beginning of the time step (Pa). As detailed in Appendix D, the parameter f depends on the axial location of the defect. If the plenum is gas-filled (bottom-end defect):

$$f = \frac{\Delta P(0)}{P_c} \frac{\xi V_p}{V_{gap}}, \quad (11)$$

while, if the rod is entirely filled with water (top-end defect):

$$f = \frac{\Delta P(0)}{\beta}, \quad (12)$$

where V_g = volume of gas in plenum ($= \xi V_p$) (m^3), V_{gap} = fuel-to-clad gap volume (m^3), V_p = plenum volume (m^3), ξ = volumetric fraction of gas in the plenum, P_c = coolant pressure (Pa), and β = fluid expansion coefficient (Pa).

The fluid expansion coefficient β can be estimated from:

$$\beta = \frac{\Delta P(0)\rho}{\Delta \rho}, \quad (13)$$

where $\Delta \rho$ is the change in density over the time step. The fluid density ρ is relatively independent of pressure. Thus, for typical shutdown transients, ρ (in kg/m^3) can be represented by a polynomial correlation in temperature T ($^{\circ}C$) over the temperature range of 50 to 320 $^{\circ}C$, based on standard steam table values:

$$\rho = 973.8 + 0.1382T - 3.266 \times 10^{-3}T^2. \quad (14)$$

Similarly, the fluid viscosity μ ($kg/m \cdot s$) is independent of pressure and can be calculated over the same temperature range from the correlation in T ($^{\circ}C$):

$$\mu = 8.312 \times 10^{-4} - 9.265 \times 10^{-6}T + 4.816 \times 10^{-8}T^2 - 1.187 \times 10^{-10}T^3 + 1.121 \times 10^{-13}T^4. \quad (15)$$

Consequently, the fraction f in Eq. (11) is much larger than that in Eq. (12) for the pressure and temperature transients experienced during shutdown.

2.2.3. Mass balance in RCS

With a knowledge of the release rate for the processes of diffusion, $R_{diff}(t)$ (Eq. (8)), and convection, $R_{conv}(t)$ (Eq. (9)), the coolant iodine inventory can be derived from the mass balance equation in the RCS (compare with Eq. (2)):

$$\frac{dN_c}{dt} = R_c(t) - (\lambda + L)N_c, \quad (16)$$

where $R_c(t) = R_{diff}(t) + R_{conv}(t)$. Eq. (16) is conservative since it assumes that both transport processes are opera-

tive. The solution of Eq. (16), subject to the initial condition that $N_c(t=0) = N_{co}$ at the beginning of the time step, is

$$N_c(t) = \left[N_{co} + N_{go} \left\{ \left(\frac{k_o}{L - k_o/f} \right) [e^{(L - k_o/f)t} - 1] + \left(\frac{k}{L - k} \right) [e^{(L - k)t} - 1] \right\} \right] e^{-(\lambda + L)t}. \quad (17)$$

In general, $k_o/f \gg L$, and Eq. (17) reduces to

$$N_c(t) = \left[N_{co} + N_{go} \left\{ f[1 - e^{-(k_o/f)t}] + \left(\frac{k}{k - L} \right) \times [1 - e^{(L - k)t}] \right\} \right] e^{-(\lambda + L)t}. \quad (18)$$

Similarly, the mass balance equation for the inventory in the gap is

$$\frac{dN_g}{dt} = -R_c(t) - \lambda N_g. \quad (19)$$

Eq. (19) only considers losses due to release to the coolant and radioactive decay. Any effect of iodine deposition onto the internal fuel/clad surfaces is conservatively ignored; however, this effect is not believed to be significant since the leaching rate constant (k) will be much greater than the corresponding deposition rate constant (see Appendix B) as evidenced, for example, in iodine transport studies at 100°C [30,31]. The solution of Eq. (19) is

$$N_g(t) = N_{go} \left\{ e^{-\lambda t} - f[1 - e^{-(k_o/f)t}] \right\} e^{-\lambda t}, \quad (20)$$

where N_{go} is the gap inventory of iodine at the beginning of the time step. Immediately after shutdown, N_{go} can be calculated from the measured coolant inventory N_{co} with the use of Eq. (7). The exponential term $\exp\{-(k_o/f)t\}$ in Eq. (20) rapidly approaches zero over the given time step. Hence, it can be seen that even if no diffusion were to occur (i.e., $k = 0$), the existing gap inventory would still be completely depleted in the given time step when f is unity.

Iodine-rich water will be expelled from the rod as a result of a forced-convective release. The volume of water displaced in the plenum will be occupied by the expanding gas. This process will continue at each time step. The volume of water ΔV displaced in a given time step is:

$$\Delta V = A_{gap} \int_0^t v(t) dt = fV_{gap} [1 - e^{-t/\tau^*}] \approx fV_{gap}, \quad (21)$$

where A_{gap} is the cross-sectional area of the gap, and τ^* ($=f/k_o$) is the characteristic time of pressure equalization that is of the order of one second at 14 MPa as shown in Section C.1. The bulk-flow velocity $v(t)$ in Eq. (C.16) has been used in the derivation of Eq. (21). Thus, at each time

step for Eq. (11), the volume of gas in the plenum ($V_g = \xi V_p$) will be increased by an amount ΔV .

2.3. I_CODE implementation

The iodine spike model of Section 2.2.3, which has been developed as the analytical solutions of the transport differential equations for diffusion and convection, has been implemented into a computer code called I_CODE (Iodine-spike CONvection and Diffusion Estimate) [32].

The I_CODE program is based on the analytical results of Eqs. (10), (11), (18), (20) and (21). This code maintains an appropriate mass balance of iodine in the fuel-to-clad gap and reactor coolant system. The initial gap inventory is calculated from the measured, steady-state coolant activity in accordance with Eq. (7). The program implicitly assumes that the defect is located at the bottom end of the rod (Section C.1). This defect representation is conservative since the release fraction f is much larger for the expanding gas in the plenum than for that associated with fluid expansion (see Section 2.2.2).

3. Model validation

The iodine-spiking model was validated with data collected from fifteen reactor trips of various PWRs. Table 1 gives the case number, plant, operating utility, the vendor that supplied the nuclear steam supply system (NSSS), and the date of data collection.

The input data needed as a function of time for the I_CODE application (Section 2.3) included: the reactor power, coolant pressure, coolant temperature and the coolant cleanup rate constant. This information, as well as the measured coolant activity for ^{131}I , are shown for several typical cases in Fig. 1. For the Cases 1 to 4, 6, 7, and 10 to 12 in Table 1, the coolant temperature data were estimated as 30°C below the coolant saturation temperature (based on the measured pressure data). For the Cases 2 to 5, 11, and 12, the cleanup rate constant (L) was obtained from the slope of a semi-log plot of the coolant activity versus time for those periods that were away from any transient events of power, pressure or temperature. The zero time was chosen to correspond to the time when the reactor power was at a value of 40% of full power or less (during the shutdown event). This value would guarantee an average fuel rod power of less than ~ 10 kW/m, i.e., in this case, the temperature of the fuel surface would be below that of coolant saturation so that only water would be present in the gap [33].

The I_CODE program was applied to the given database. A time-step size of one hour was chosen for the simulation of the entire database. In this calculation, a defect at the bottom end of the rod (Section 2.3) was

assumed, i.e., the plenum volume (V_p), the fuel-to-clad gap volume (V_{gap}), and the fuel stack length (l) were based on the nominal fabrication values listed in Table 2. The fitting parameters of the model included: ν , k and ξ . The parameter ν provides an estimate of the iodine inventory in the gap that is available for release, given the measured value of the steady-state inventory in the coolant (see Eq. (7)). The parameter k describes the rate of diffusional transport of the water-soluble iodine in the gap following reactor shutdown. Finally, ξ corresponds to the fraction of the plenum volume filled with gas, i.e., this parameter determines the quantity of iodine-rich water that can be expelled as result of the expanding plenum gas with pressure or temperature transients. The values of the fitting parameters for each case are given in Table 2.

The measured and predicted values of the ^{131}I coolant activity concentration as a function of time are shown for the selected cases in Fig. 1. The complete analysis for the total database is provided in Ref. [32]. In general, the model is in good agreement with the measured data. For example, Fig. 1(a) displays a case where the coolant temperature had to be estimated; the model, however, is able to capture the second iodine spike that results from a further depressurization. Fig. 1(b) is an example where there is no coolant depressurization, so that only a diffusional release occurs. On the other hand, Fig. 1(c) is representative of a typical shutdown where there is both a pressure and temperature change as a result of normal cooldown operation. Finally, Fig. 1(d) shows an iodine-spike event where three of the short-lived isotopes of iodine have been monitored. In all circumstances, the model is able to reproduce the spiking behavior.

The respective fitted parameters in Table 2 typically

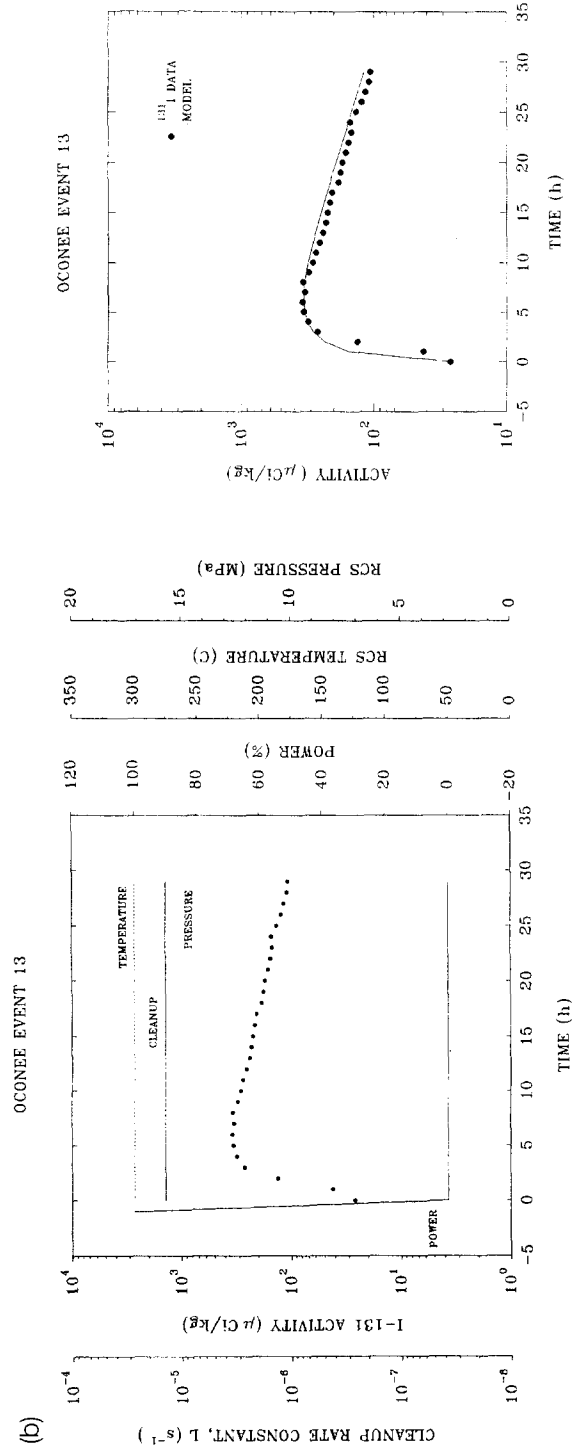
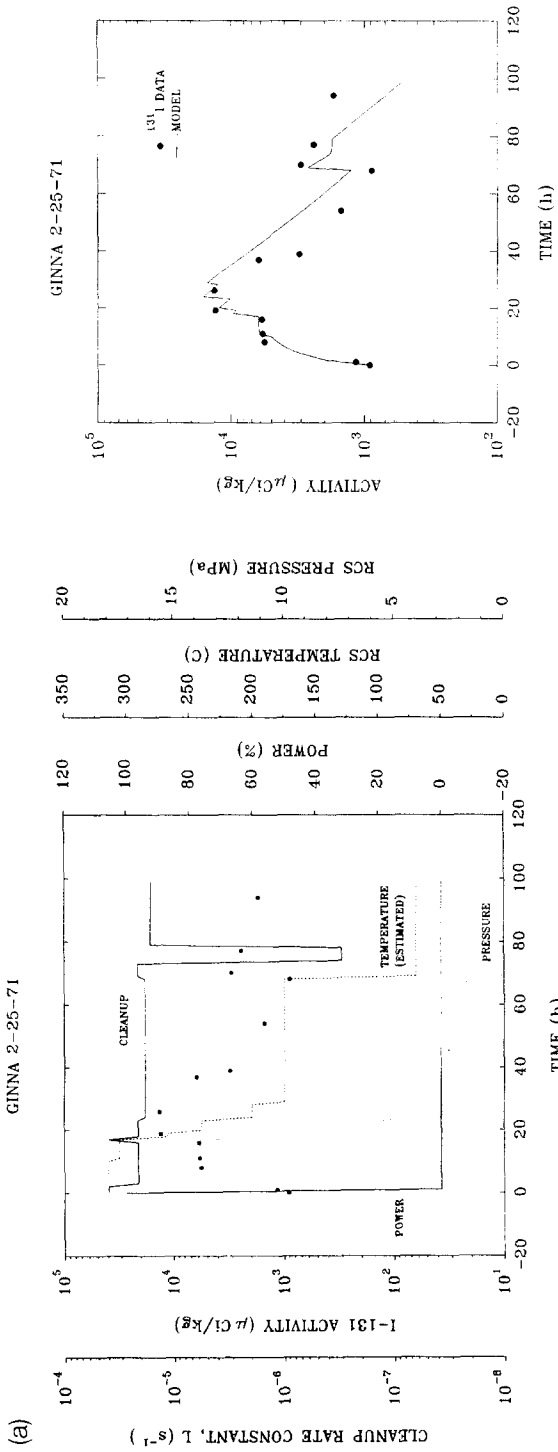
range over an order of magnitude. These values, however, are in good agreement with other studies. For instance, for defective PWR fuel, the parameter ν has been estimated by Beyer [6] in the range of 4×10^{-7} to $1 \times 10^{-5} \text{ s}^{-1}$, while Bishop [14] has recommended a 'typical' value of $5 \times 10^{-6} \text{ s}^{-1}$. In addition, based on an experimental in-reactor program with defective CANDU fuel rods, Lewis has obtained a value of ν in the range of 6×10^{-7} to $2 \times 10^{-5} \text{ s}^{-1}$, and a value of k equal to $4 \times 10^{-5} \text{ s}^{-1}$ for a single defect test [3,19]. For the Three Mile Island-2 plant (case 15), the fitting parameters for the iodine isotopes (^{131}I , ^{133}I and ^{135}I) are also self-consistent, and provide a good prediction of the release behavior (see Fig. 1(d)).

This range in values reflects the different physical characteristics of the defective rods (i.e., defect size and location) (see Section 2). For example, a small value of ν ($\nu = 1.2 \times 10^{-7} \text{ s}^{-1}$) represents a very 'tight' defect whereas a large value would represent a severely-hydrided failure [10]. Small defects due to debris fretting, for example, have been observed in the Haddam Neck fuel which has used stainless steel cladding that is not susceptible to secondary hydriding [34]. The value of k is particularly sensitive to the migration-path length (see Appendix A) where the iodine on shutdown becomes much more mobile with less chemical trapping as it ionically diffuses in the liquid medium. On the other hand, ξ will be less affected by the defect size but will depend more on the axial defect location (relative to that of the plenum), as well as on the fuel rod power and burnup. The range of values for ξ represents the possible variation of the stored inventory of non-condensable gases present in the plenum on shutdown. As seen in Table 2, this parameter varies somewhat less than the other two (i.e., typically from 0.1 to 0.3) since the

Table 1
PWR database used for model validation

Case	Plant	Utility	NSSS Vendor ^a	Date
1	GINNA	Rochester Gas and Electric Corp.	W	2-25-71
2	GINNA	Rochester Gas and Electric Corp.	W	4-13-72
3	Haddam Neck	Connecticut Yankee Atomic Power Co.	W	6-10-72
4	Haddam Neck	Connecticut Yankee Atomic Power Co.	W	4-15-71
5	McGuire-1	Duke Power Co.	W	1-23-94
6	Mihama	Kansai Electric Power Co., Inc.	W	5-25-74
7	Mihama	Kansai Electric Power Co., Inc.	W	1-8-75
8	Oconee	Duke Power Co.	B & W	—
9	Point Beach	Wisconsin Electric Power Co.	W	2-26-76
10	Point Beach	Wisconsin Electric Power Co.	W	4-4-74
11	San Onofre	Southern California Edison Co.	C-E	12-26-72
12	San Onofre	Southern California Edison Co.	C-E	10-2-70
13	Surry-1	Virginia Power	W	10-17-76
14	Surry-1	Virginia Power	W	8-13-76
15	Three Mile Island-2	GPU Nuclear Corp.	B & W	2-20-76

^a W = Westinghouse; B & W = Babcock and Wilcox; C-E = Combustion Engineering.



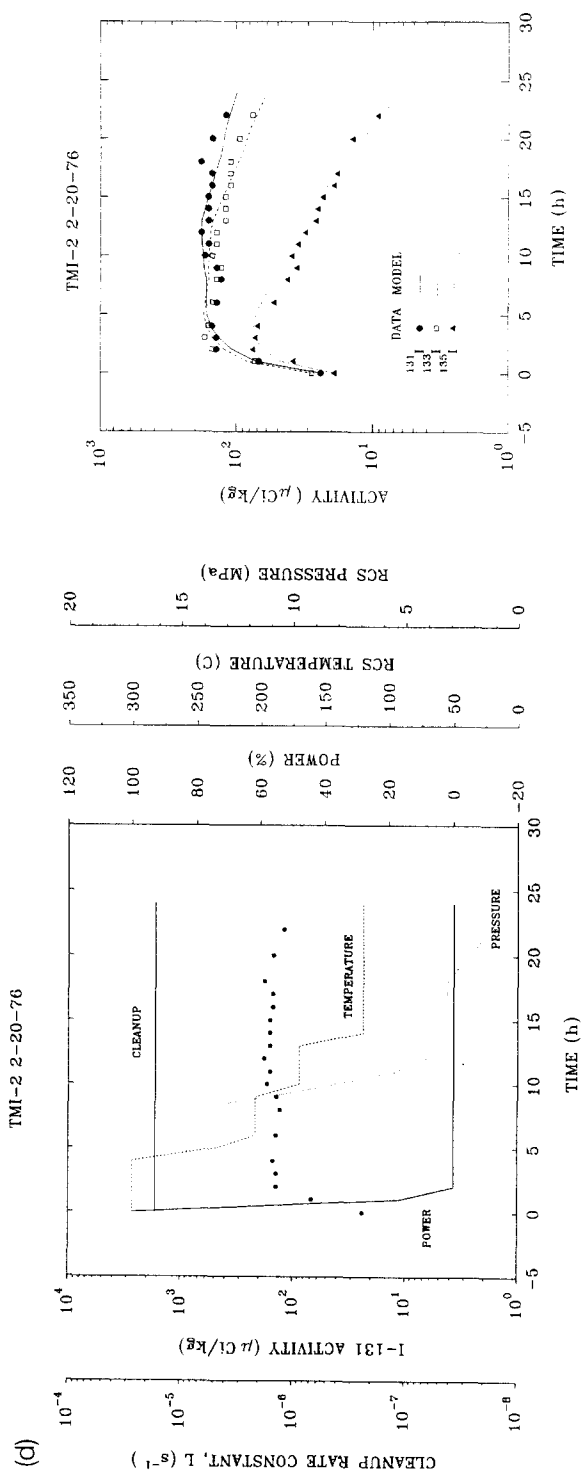
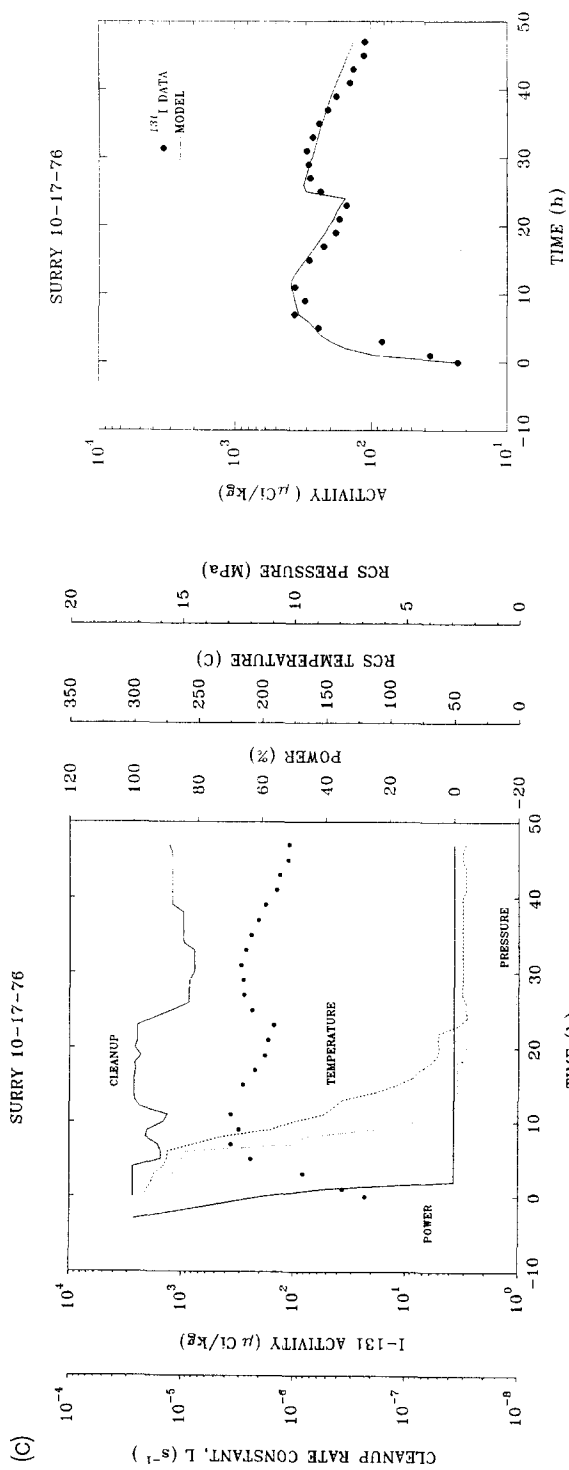


Table 2
Parametric values used for model validation

A. Fabrication parameters					
Fuel stack length, l (m)		3.6			
Plenum volume, V_p (m ³)		1.5×10^{-5}			
Fuel-to-clad gap thickness, h (μm)		95			
Pellet diameter, D_{pellet} (m) ^a		9.3×10^{-3}			
B. Model parameters					
case	plant	Fixed value	Fitted parameters		
		measured steady-state coolant activity, C_{co} ($\mu\text{Ci}/\text{kg}$)	steady-state escape rate constant, ν (s ⁻¹)	transient escape rate constant, k (s ⁻¹) ^b	volumetric fraction of gas in plenum, ξ
1	Ginna	910	9.7×10^{-7}	5.5×10^{-6}	0.2
2	Ginna	700	3.6×10^{-7}	1.4×10^{-5}	0.2
3	Haddam Neck	483	8.1×10^{-7}	1.1×10^{-5}	0.2
4	Haddam Neck	35	1.2×10^{-7}	5.5×10^{-6}	0.1
5	McGuire-1	14.5	1.0×10^{-6}	1.4×10^{-5}	0.3
6	Mihama	100	2.9×10^{-6}	2.8×10^{-5}	0.1
7	Mihama	50	6.6×10^{-7}	1.4×10^{-5}	0.3
8	Oconee	26.3	8.6×10^{-7}	9.4×10^{-5}	N/A
9	Point Beach	6.1	1.5×10^{-6}	2.2×10^{-4}	N/A
10	Point Beach	127	4.0×10^{-7}	5.5×10^{-6}	0.2
11	San Onofre	23	3.2×10^{-7}	5.5×10^{-6}	0.06
12	San Onofre	124	8.6×10^{-7}	7.9×10^{-6}	0.02
13	Surry-1	23	7.3×10^{-7}	2.8×10^{-5}	0.1
14	Surry-1	33.1	7.3×10^{-7}	3.2×10^{-5}	0.2
15	Three Mile Island-2 ^{c,d}	24	1.4×10^{-6}	5.0×10^{-5}	0.3
Average	179	9.1×10^{-7}	3.6×10^{-5}	0.18	

N/A = not applicable (no change in pressure/temperature occurred).

^a The gap volume is calculated as $V_{\text{gap}} = \pi D_{\text{pellet}} h l$.

^b Value quoted for a temperature and pressure of 300°C and 15 MPa, respectively. The value of k for other temperatures and pressures can be calculated from Eq. (A.10).

^c Value of measured steady-state coolant activity for ¹³¹I. The values for the other isotopes are: 28 $\mu\text{Ci}/\text{kg}$ (I-133) and 19 $\mu\text{Ci}/\text{kg}$ (I-135).

^d Value of ν quoted for ¹³¹I. For the other isotopes, $\nu_{\text{I-133}} = 1.9 \times 10^{-6} \text{ s}^{-1}$ and $\nu_{\text{I-135}} = 4.0 \times 10^{-6} \text{ s}^{-1}$. All three isotopes have the same value of k and ξ .

transport of non-condensable gases is not a chemically-affected process [3].

4. SGTR/MSLB transient analysis

4.1. Comparison with SRP methodology

The more physical model developed in Section 2 can be used to evaluate the standard review plan (SRP) guideline [21]. In the present analysis, a steam generator tube

rupture (SGTR) accident is assumed to occur with coincident iodine spike. A typical event sequence for a main steam line break (MSLB) is shown in Fig. 2 assuming a leak rate of 100 gpm [35]. For this calculation, the pressure and temperature histories of Fig. 2 are employed, where a time-step size of 3.75 min was chosen to accurately represent these data. In accordance with the assumptions of the model, the coolant temperature is always below the saturation temperature for the MSLB event. The pressure transient in Fig. 2, however, is more rapid than that experienced during the normal shutdown procedure (compare with Fig. 1). The bulk velocity in the fuel-to-clad gap

Fig. 1. Representative reactor shutdown events. The reactor power, and pressure, temperature, cleanup rate constant and ¹³¹I activity concentration for the RCS, are shown in the left figure, and a comparison of the measured and predicted ¹³¹I coolant activity concentration history is shown in the right figure. (a) Case 1 (Ginna 2–25–71), (b) Case 8 (Oconee event 13), (c) Case 13 (Surry 10–17–76), (d) Case 15 (TMI-2 2–20–76).

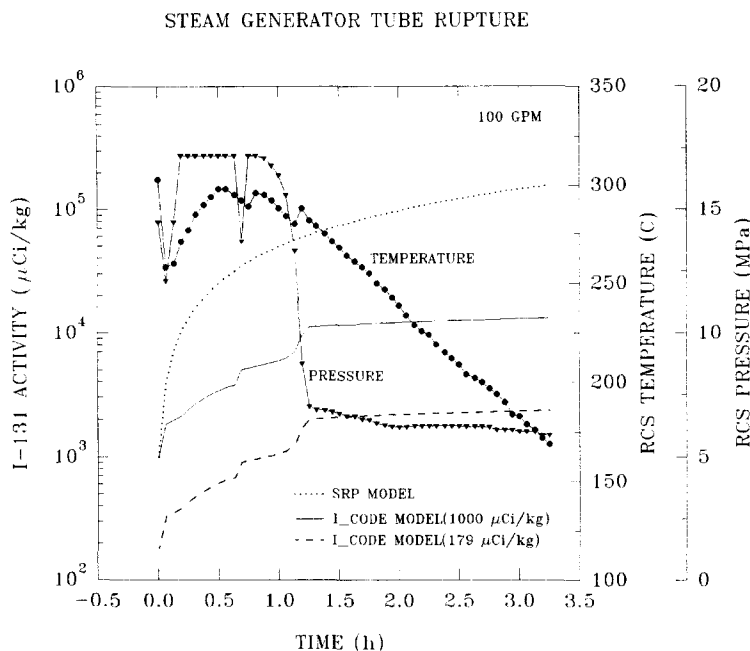


Fig. 2. Comparison of SRP and diffusion/convection model predictions of the ^{131}I coolant activity concentration for a SGTR/MSLB sequence (leakage rate of 100 gpm). The RCS pressure and temperature histories for the event are also shown.

(averaged over a given time step) can be calculated for the present transient using Eq. (C.16),

$$\bar{v} = \int_0^t v(t) dt / \int_0^t dt = \frac{\gamma t^*}{t} [1 - e^{-t/t^*}], \quad (22)$$

where $\gamma = k_0 l$ and $t^* = 12 \mu l^2 V_g / (h^2 P_c V_{\text{gap}})$ is the characteristic time constant for pressure equalization in the plenum (see Appendix C). For example, assuming the nominal values in Section C.1 and $\Delta P(0) \approx 2 \times 10^5$ Pa in Eq. (10), \bar{v} is evaluated as 6.5×10^{-3} m/s. A calculation of the Reynold's number [25] ($\text{Re} = 2 h \bar{v} \rho / \mu = 5.2$, where ρ is the fluid density $= 7.2 \times 10^2$ kg/m³ evaluated in Eq. (14)) indicates the presence of laminar flow, as assumed in the derivation of the pressure-differential release model.

Following the analysis of Postma [20], the SRP guidelines can be quantified for an individual plant. It is assumed that a reactor trip occurs instantaneously from 100% of full power at time zero, and that the coolant cleanup system does not operate for time greater than zero (i.e., credit is only taken for radioactive decay). The SRP guideline stipulates that for an accident-initiated spike, a release rate of 500 times the corresponding value at equilibrium must be used; the equilibrium release rate R_{co} (atoms/s) is based on a coolant concentration level of $C_{\text{co}} = 1000 \mu\text{Ci/kg}$, i.e., from Eq. (2) [21]:

$$R_{\text{co}} = \frac{(\lambda + L_0)}{\lambda} M C_{\text{co}} \epsilon, \quad (23)$$

where λ = decay constant (s^{-1}), L_0 = steady-state coolant cleanup rate constant (s^{-1}), M = RCS mass (kg), and ϵ = conversion factor ($= 3.7 \times 10^4$ Bq/ μCi). A typical

value of L_0 is $2 \times 10^{-5} \text{ s}^{-1}$ [20]. The transient release rate is therefore taken to be a constant such that $R_c = 500 R_{\text{co}}$. The time-dependent coolant activity concentration for the spike event, $C_c(t)$ (in $\mu\text{Ci/kg}$), follows from the solution of the mass-balance equation (see Eq. (16)) where L is now equal to zero:

$$N_c(t) = N_{\text{co}} e^{-\lambda t} + \frac{R_c}{\lambda} (1 - e^{-\lambda t}), \quad (24)$$

or equivalently,

$$C_c(t) = C_{\text{co}} \left\{ e^{-\lambda t} + 500 \left(\frac{\lambda + L_0}{\lambda} \right) [1 - e^{-\lambda t}] \right\}. \quad (25)$$

The SRP model prediction of Eq. (25) is shown in Fig. 2 for ^{131}I . This calculation can be compared to the model prediction of Section 2.2. The gap inventory can be conservatively estimated from the assumed SRP value of $C_{\text{co}} = 1000 \mu\text{Ci/kg}$ and an average value of $\nu = 9.1 \times 10^{-7} \text{ s}^{-1}$ in Table 2¹. Thus, using the I_CODE program,

¹ The use of an 'average' value is in fact more representative since the smallest value of ν (implying a 'tight' defect) would require an anomalously large number of defects (i.e., hundreds of failed rods) in order to produce a large coolant activity level of $1000 \mu\text{Ci/kg}$. To provide a significant iodine release during the shutdown event, a worst case analysis could consider a small value of ν , and a large value for the leaching rate constant k , in accordance with the given range of values in Table 2. This combination of values is again unphysical since a small value of ν points to a small defect and hence a small value of k as well.

as the gap inventory is depleted the predicted coolant activity levels, in contrast to the SRP model, approach a relatively constant level because of a depleted gap inventory and little radioactive decay in the coolant (assuming no coolant cleanup). A more realistic estimate can be further obtained with the I_CODE model using an average value of the steady-state coolant concentration for the

fifteen cases in Table 2 (i.e., 179 $\mu\text{Ci}/\text{kg}$) [32]. Thus, at two hours, it can be seen that the SRP analysis is overly conservative by a factor of 45.

4.2. Comparison with actual SGTR events

The I_CODE model can be validated against the observed iodine-spiking behavior in actual SGTR events at

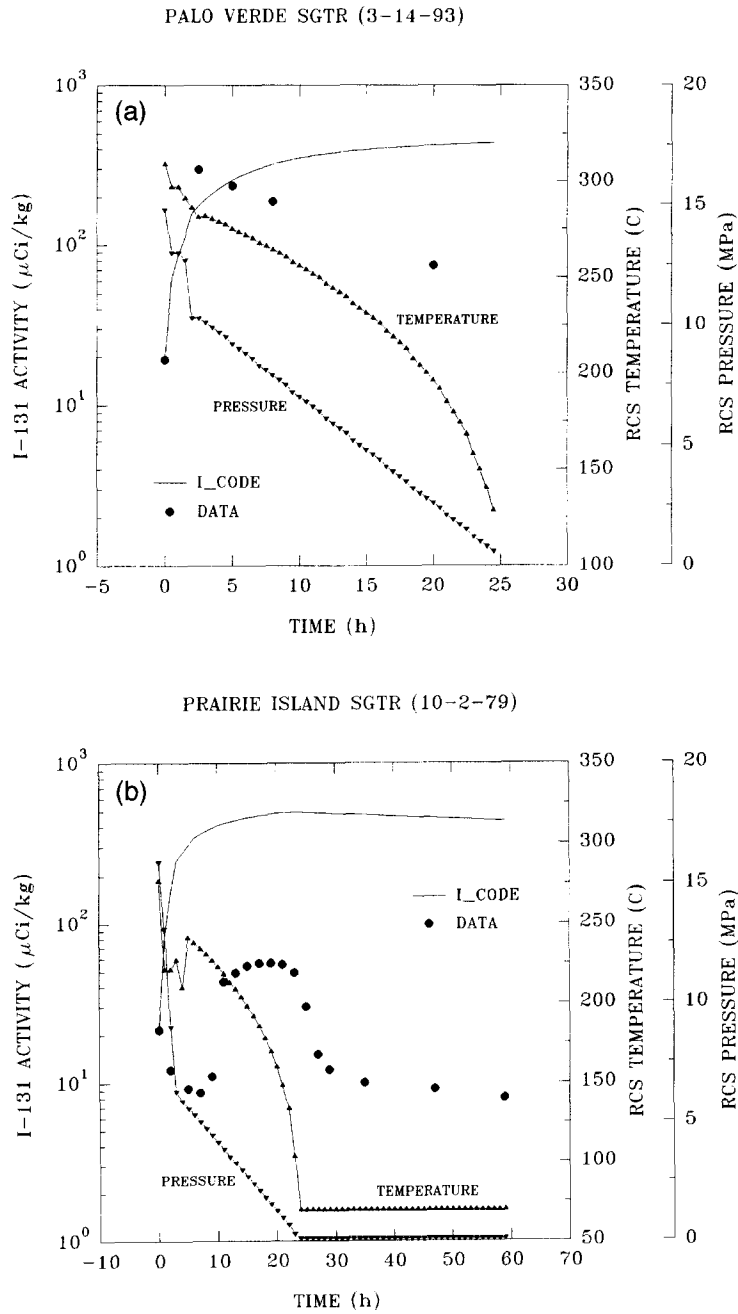


Fig. 3. Comparison of the measured and predicted I_CODE ¹³¹I spike activity for the (a) Palo Verde SGTR, (b) Prairie Island SGTR. The input RCS pressure and temperature histories are also shown.

the Palo Verde and Prairie Island stations [36]. The activity in the fuel-to-clad gap can be estimated from the measured activity in the coolant using Eq. (7), and a coolant cleanup rate constant of $L_0 \approx 2 \times 10^{-5} \text{ s}^{-1}$, for the steady-state situation. In accordance with the methodology of Section 4.1, L is assumed to be zero after the reactor trip. In addition, the average fitted coefficients in Table 2 were used for the I_CODE calculation. A comparison of the I_CODE predictions with the measured coolant activities are shown in Fig. 3(a) and (b), along with the RCS pressure and temperature histories. Since temperature data were not available after 2.5 and 4 h for the Palo Verde and Prairie Island cases, respectively, this parameter was estimated from the coolant saturation temperature (see Section 3). In the present analysis, time zero corresponds to the time of reactor trip.

Early in the Palo Verde transient (see Fig. 3(a)), the I_CODE prediction is in good agreement with the measured results; however, later in the transient, it overpredicts the measured data as a result of the conservative approach taken where any losses in the system were neglected. For example, losses may arise either due to leakage of primary coolant to the secondary side, a dilution of the RCS by safety injection and the inflowing of secondary water, or operation of the coolant cleanup system (which is normally turned off after reactor trip). The I_CODE model overpredicts the measured data in the Prairie Island case (see Fig. 3(b)), typically by an order of magnitude. The observed delay in the iodine spike following reactor shutdown in Fig. 3(b), may be possibly attributed to a reduced transport of iodine to the sampling station. For instance, the main coolant pumps were turned off about two minutes after reactor trip, whereas the pump in the unaffected loop B was restarted about seven hours after the beginning of the event, which coincides with the time when the iodine activity is first observed to increase [36]. The initial drop in iodine activity may result from the effect of coolant leakage, dilution or coolant cleanup, which is conservatively ignored in the I_CODE simulation.

In comparison, the SRP prediction shown by the dotted line in Fig. 2 can be directly applied and, as expected, significantly over-predicts the observed release behavior in Fig. 3(a) and (b).

In conclusion, a best-estimate analysis with the I_CODE model appears to represent or over-predict the measured spike event for the two SGTR cases, in accordance with the methodology of Section 4.1. On the other hand, the SRP model is highly conservative for these two events.

5. Conclusions

(1) A more physically-based model has been developed to describe the iodine-spiking phenomenon for PWR fuel on reactor shutdown. The model considers the total mass balance of iodine in both the fuel-to-clad gap and reactor

coolant system (RCS). The transport of iodine in the gap and its subsequent release to the coolant is treated by diffusion theory. In addition, the model considers the convective release for a pressure differential between the interior of the rod and the bulk coolant that can result with variable pressure and temperature conditions in the RCS during the shutdown event.

(2) The model has been successfully validated against a database of iodine-spiking events that have occurred during normal PWR operation, and for two steam generator tube rupture events.

(3) The model has been used to evaluate the standard review plan (SRP) methodology for an iodine spike initiated by a steam generator tube rupture and main steam line break sequence. A best-estimate calculation with the model indicates that the SRP methodology is conservative by at least an order of magnitude. This result is to be expected since the SRP methodology ignores any mass conservation in the gap and assumes a constant release rate. In contrast, the I_CODE model: (i) maintains a mass balance in both the gap and coolant, where there is only a finite supply of iodine in the gap which is continually being depleted, and (ii) employs an exponentially-decreasing release rate (in comparison to a constant release rate in the SRP model), in accordance with physical transport processes of diffusion and convection.

Acknowledgements

This work was sponsored by the Electric Power Research Institute.

Appendix A. Derivation of diffusion model

The iodine-spike process can be described by a diffusion mechanism that is governed by the transport equation:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} - \lambda C \quad (\text{A.1})$$

where x = axial direction along the fuel-to-sheath gap (m), t = time (s), C = concentration of solute free to diffuse in the gap (Bq/m), D = diffusion coefficient of iodine in the gap (m^2/s), and λ = radioactive decay constant (s^{-1}). The plenum is expected to contain some non-condensable gases on shutdown. Hence, it is assumed that only iodine diffusion occurs in the water-filled gap, i.e., any diffusion in the water-filled part of the plenum is neglected.

Consider the solution of Eq. (A.1) for a defect located at the mid-axial location of the fuel stack of length l . Initially, the concentration of the diffusing substance is zero at the defect site, but equal to a finite value C_0 far away from the defect location. At the ends of the fuel stack there is a reflexive (impenetrable) boundary, while at the defect site the concentration must remain zero since iodine

is being continually swept away by the coolant. These conditions are mathematically described by:

$$C = C_0 \sin \frac{\pi x}{l}, \quad 0 < x < l/2, \quad t = 0 \quad (\text{A.2})$$

$$C = 0, \quad x = 0, \quad t > 0 \quad (\text{A.3})$$

$$\frac{\partial C}{\partial x} = 0, \quad x = l/2, \quad t > 0. \quad (\text{A.4})$$

The sinusoidal distribution in Eq. (A.2) is a reasonable approximation to the actual profile developed during the steady-state situation with steam present in the gap [3].

The solution of Eq. (A.1) can be obtained by the method of separation of variables [15,19]:

$$C(x, t) = C_0 \sin \frac{\pi |x|}{l} \exp\{-(\lambda + \pi^2 D/l^2)t\},$$

$$-\frac{l}{2} < x < \frac{l}{2}. \quad (\text{A.5})$$

The rate at which iodine leaves the defect is given by the Fick's law of diffusion:

$$R(t) = 2D \left. \frac{\partial C}{\partial x} \right|_{x=0} = \frac{2\pi DC_0}{l} \exp\{-(\lambda + \pi^2 D/l^2)t\}, \quad (\text{A.6})$$

where the factor of two accounts for release from both halves of the element. The total number of atoms in the fuel-to-sheath gap available for release at time $t = 0$ is

$$N_{g0} = 2 \int_0^{l/2} C(x, 0) dx = 2C_0 \int_0^{l/2} \sin \frac{\pi x}{l} dx = \frac{2C_0 l}{\pi}. \quad (\text{A.7})$$

Therefore substituting Eq. (A.7) into Eq. (A.6) yields

$$R(t) = kN_{g0} \exp\{-(\lambda + k)t\}, \quad \text{where } k = \frac{\pi^2 D}{l^2}. \quad (\text{A.8})$$

Eq. (A.8) is identical to that derived by diffusion theory where the initial concentration profile is assumed to have a more general form [15]. The above analysis can be easily generalized for a rod containing n -multiple defect sites located symmetrically along the clad [15]. In this case, the rate constant k has the more general form:

$$k = n^2 \pi^2 D/l^2. \quad (\text{A.9})$$

It also follows that $n = 1$ for a mid-length defect (see Eq. (A.8)), and $n = 1/2$ for a single defect located at one end of the rod [15]. As shown in Appendix B, the release-rate expression in Eq. (A.8) can be derived from first-order kinetic theory as well.

A correction factor can be applied to Eq. (A.9) to account for the effect of a temperature or pressure change on the diffusivity. Using the Nernst theory to describe the

ionic diffusivity, and the Piszarskii–Walden relation to relate the ionic conductivity to the fluid viscosity μ , k has the following dependence on temperature (T in K) and pressure (P) [15]:

$$k \propto \frac{T}{\mu} \quad \text{or} \quad \frac{k(T_1, P_1)}{k(T_2, P_2)} = \frac{T_1 \mu(T_2, P_2)}{T_2 \mu(T_1, P_1)}. \quad (\text{A.10})$$

The fluid viscosity is generally insensitive to pressure. However, with decreasing temperature, the rate constant k (and diffusivity D) will decrease since the fluid will become more viscous.

Appendix B. Derivation of first-order kinetic model

The iodine-spike release can be modelled as a first-order rate process, where the release rate R is proportional to the total number of atoms of solute in the gap (N_g):

$$R = kN_g, \quad (\text{B.1})$$

where k is a first-order rate constant. The mass balance in the fuel-to-clad gap is

$$\frac{dN_g}{dt} = -kN_g - \lambda N_g. \quad (\text{B.2})$$

For the initial condition that $N_g(t=0) = N_{g0}$, the solution of Eq. (B.2) is

$$N_g(t) = N_{g0} \exp\{-(\lambda + k)t\}. \quad (\text{B.3})$$

Using Eqs. (B.1) and (B.3), the release rate for this simple kinetic model is

$$R(t) = kN_{g0} \exp\{-(\lambda + k)t\}. \quad (\text{B.4})$$

Eq. (B.4) is identical to the release-rate expression in Eq. (A.8). However, the main advantage of the earlier diffusion approach is that the rate constant k can be described in terms of the axial defect location and ionic diffusivity (see Eq. (A.9)).

Appendix C. Plenum pressure equalization

C.1. Defect at bottom end of rod

The time required for the pressure to equalize in the plenum can be calculated with an expanding gas-piston model, where it is assumed that a single defect is located at the bottom end of the fuel rod. After shutdown, part of the plenum will be filled with gas, while the remainder of the internal void will contain water. The volume of gas (V_g) and water (V_w) in the plenum is equal to some fraction of the total plenum volume (V_p), such that

$$V_g = \xi V_p, \quad V_w = (1 - \xi) V_p. \quad (\text{C.1})$$

If the primary coolant depressurizes, or there is an increase in the system temperature, any gas trapped in the plenum will expand forcing water out of the fuel rod until an equilibrium is reached between the plenum and RCS pressure. As the gas expands, the volume of water in the plenum will decrease such that

$$\frac{dV_g}{dt} = -\frac{dV_w}{dt} \quad (C.2)$$

From the mass balance of water in the gap

$$\frac{dV_w}{dt} = -vA_{gap} \quad (C.3)$$

where A_{gap} is the constant gap cross-sectional area and v is the fluid velocity in the gap. In this analysis, it is assumed that there is a negligible pressure drop across the defect. In the case of laminar, incompressible flow in the gap, the annular-flow velocity v can be obtained from the pressure gradient via a Hagen–Poiseuille law [25]

$$v = -\frac{R^2}{8\mu} \left\{ 1 + \kappa^2 - \frac{1 - \kappa^2}{\ln(1/\kappa)} \right\} \frac{dP}{dx} \quad (C.4)$$

where μ is the dynamic water viscosity, $\kappa = 1 - h/R$, R is the inside radius of the clad, and h is the gap thickness. If $h < R$, Eq. (C.4) reduces to the standard law for two parallel planes separated by a distance h [26,27]:

$$v = -\frac{h^2}{12\mu} \frac{dP}{dx} = \frac{h^2}{12\mu} \frac{\Delta P}{l} \quad (C.5)$$

In the derivation of the second term of Eq. (C.5), it is assumed that the pressure differential,

$$\Delta P(t) = P_p(t) - P_c \quad (C.6)$$

occurs over the total fuel stack length l , where $P_p(t)$ is the plenum gas pressure and P_c is the coolant pressure (that is taken to be constant over a given time step). Eq. (C.5) is identical to that derived by Olander and Vaknin using the more general treatment of McCabe and Smith [28], except that the constant 12 is replaced by 32 [24]. This discrepancy presumably results from the incorrect assumption of pipe flow rather than annular flow in the latter analysis of Olander and Vaknin.

Since the effective mass of gas in the plenum, m_g , is constant (i.e., due to a gas-bottle effect), Eqs. (C.2) and (C.3) yield

$$\frac{d(1/\rho_g)}{dt} = \frac{vA_{gap}}{m_g} \quad (C.7)$$

where $\rho_g = m_g/V_g$. The ideal gas law for the plenum is

$$P_p = \rho_g RT/M_g = n_g RT/V_g \quad (C.8)$$

where R is the ideal gas constant, T is the temperature, M_g is the molecular weight of the gas, and n_g is the

number of moles of gas. Hence, under constant temperature conditions, Eq. (C.7) becomes

$$\frac{d(1/P_p)}{dt} = \frac{vA_{gap}}{P_p V_g} \quad (C.9)$$

Using the chain rule for the derivative, Eq. (C.9) can be rewritten as

$$\frac{d(P_p)}{dt} = -\frac{vA_{gap} P_p}{V_g} \quad (C.10)$$

Hence, using Eqs. (C.5) and (C.6), Eq. (C.10) becomes

$$\frac{d(\Delta P)}{dt} = -\frac{h^2}{12\mu} \frac{\Delta P}{l^2} \frac{V_{gap}}{V_g} P_p \quad (C.11)$$

where $V_{gap} = A_{gap}l$. Following the analysis of Olander and Vaknin, the plenum pressure can be written as [24]

$$P_p = P_c + \Delta P = P_c \left[1 + \frac{\Delta P}{P_c} \right] \quad (C.12)$$

Assuming that $\Delta P < P_c$ for the given time step, Eqs. (C.11) and (C.12) yield

$$\frac{d(\Delta P)}{d(t/t^*)} = -\Delta P \quad (C.13)$$

where t^* is the characteristic time constant to equalization:

$$t^* = \frac{12\mu l^2}{h^2 P_c} \frac{V_g}{V_{gap}} \quad (C.14)$$

Eq. (C.14) is identical in form to that obtained in Ref. [24]. Typical values of the parameters in Eq. (C.14) are:

T	$= 300^\circ\text{C}$
P_c	$= 14 \text{ MPa} = 1.4 \times 10^7 \text{ Pa}$
μ	$= 9.0 \times 10^{-5} \text{ kg/m} \cdot \text{s}$
l	$= 3.6 \text{ m}$
h	$= 50 \text{ } \mu\text{m} = 5.0 \times 10^{-5} \text{ m}$
D_{pellet}	$= \text{pellet diameter} = 9.3 \times 10^{-3} \text{ m}$
V_g	$\approx V_p = 1.5 \times 10^{-5} \text{ m}^3$
V_{gap}	$= \pi D_{\text{pellet}} l h = 5.3 \times 10^{-6} \text{ m}^3$

Hence, using these values, $t^* \approx 1 \text{ s}$.

The solution of Eq. (C.13) is therefore given by

$$\Delta P(t) = \Delta P(0)e^{-t/t^*} \quad (C.15)$$

where $\Delta P(0)$ is the pressure gradient at $t = 0$. Using Eqs. (C.5) and (C.15), the velocity during pressure equalization is given by the time-dependent function

$$v(t) = \gamma e^{-t/t^*} \quad (C.16)$$

where $\gamma = h^2 \Delta P(0)/(12\mu l)$.

If the system pressure remains constant, but the system

temperature increases, the plenum pressure P_p will increase in accordance with the ideal gas law of Eq. (C.8). Thus, a pressure differential will result and this case can also be treated with the present methodology.

C.2. Defect at top end of rod

If a defect is located at the top of the rod, i.e., in the plenum region, the non-condensable gases can escape from the plenum. The rod will therefore be totally filled with water on shutdown. During coolant depressurization, liquid will be expelled from the rod as a result of liquid expansion from the fluid density change. If there is again no significant pressure differential across the defect, the plenum will be in equilibrium with the RCS so that a pressure gradient will only result over the axial length of the gap.

The mass balance for the fluid in the gap is given by the rate equation:

$$\frac{d(\rho V_{\text{gap}})}{dt} = -GA_{\text{gap}}, \quad (\text{C.17})$$

where ρ is the fluid density (kg/m^3), and $G (= \rho v)$ is the mass velocity of the fluid ($\text{kg}/\text{m}^2 \cdot \text{s}$). Using the chain rule for Eq. (C.17), with a constant gap volume, yields

$$\frac{dP_A}{dt} = -\frac{GA_{\text{gap}}}{V_{\text{gap}}} \frac{dP_A}{d\rho}, \quad (\text{C.18})$$

where P_A is the pressure at the bottom (intact) end of the rod. The derivative $dP_A/d\rho$ can be related to the coefficient of expansion β (in Pa) for the liquid, where [29]

$$\frac{dP_A}{d\rho} = \frac{\beta}{\rho}. \quad (\text{C.19})$$

Using Eq. (C.19), and the Hagen–Poiseuille flow velocity in Eq. (C.5) for the pressure differential

$$\Delta P(t) = P_A(t) - P_c, \quad (\text{C.20})$$

Eq. (C.18) reduces to the form of Eq. (C.13). In this case, however, the characteristic time constant is defined as

$$t^* = \frac{12\mu l^2}{h^2\beta}. \quad (\text{C.21})$$

A typical value of β at 14 MPa and 300°C is 3.6×10^8 Pa; hence, the time constant t^* is several orders of magnitude smaller than that for the gas-plenum case.

Appendix D. Derivation of convection model

In the case that there is a pressure differential (see Appendix C), the iodine release can be described by forced-convective transport [26]:

$$\frac{\partial C}{\partial t} = -v(t) \frac{\partial C}{\partial x} - \lambda C, \quad (\text{D.1})$$

where $x =$ axial direction along the fuel-to-sheath gap (m), $t =$ time (s), $C =$ concentration of solute in the gap (Bq/m), $v =$ bulk-flow velocity in the gap from pressure differential (m/s), and $\lambda =$ radioactive decay constant (s^{-1}). The time-dependent, bulk-flow velocity is given by Eq. (C.16). For a defect located at one end of the rod, the time constant t^* is given by either Eq. (C.14) or Eq. (C.21). A constant concentration profile C_0 can be assumed initially. It is further assumed for the boundary condition that there is no flow at the intact end of the rod (i.e., at $x = 0$). These conditions are mathematically described by [26]:

$$C = C_0, \quad 0 < x < l, \quad t = 0 \quad (\text{D.2})$$

$$vC = 0, \quad x = 0, \quad t > 0. \quad (\text{D.3})$$

Eq. (D.3) implies that $C = 0$ at $x = 0$ since the flow velocity is taken to be independent of position and finite throughout the axial length of the rod. Given the transformation

$$\tau = \int_0^t v(t) dt, \quad (\text{D.4})$$

Eq. (C.16) becomes

$$v(t) = \gamma - \tau/t^*, \quad (\text{D.5})$$

and Eq. (D.1) can be written as

$$\frac{\partial C}{\partial \tau} = -\frac{\partial C}{\partial x} - \left(\frac{\lambda}{\gamma - \tau/t^*} \right) C. \quad (\text{D.6})$$

Taking the Laplace transform of Eq. (D.6) with respect to the variable x , such that $c(s, \tau) = \mathcal{L}\{C(x, \tau)\}$, one obtains

$$\frac{dc}{d\tau} = -\left(s + \frac{\lambda}{\gamma - \tau/t^*} \right) c, \quad (\text{D.7})$$

where the boundary condition in Eq. (D.3) has been applied. Using the transformed initial condition, $c(s, 0) = C_0/s$, the solution of Eq. (D.7) is given as:

$$c(s, \tau) = C_0 \left[1 - \frac{\tau}{\gamma t^*} \right]^{\lambda t^*} \left\{ \frac{e^{-s\tau}}{s} \right\}. \quad (\text{D.8})$$

The inverse transform of Eq. (D.8) is

$$C(x, \tau) = C_0 \left[1 - \frac{\tau}{\gamma t^*} \right]^{\lambda t^*}, \quad \tau \leq x. \quad (\text{D.9})$$

The release rate from the defect site at the end of the rod of stack length l is

$$R(t) = v(t) C(x, t)_{x=l}. \quad (\text{D.10})$$

Using Eqs. (C.16), (D.5) and (D.9), with the relation that $C_0 = N_{\text{go}}/l$, Eq. (D.10) becomes

$$R(t) = \gamma C_0 \exp\{-(\lambda + 1/t^*)t\} = k_0 N_{\text{go}} \times \exp\{-(\lambda + f^{-1}k_0)t\}, \quad (\text{D.11})$$

where $k_o = (h^2 \Delta P(0)) / (12 \mu l^2)$. The parameter f depends on whether the plenum is gas filled,

$$f = \frac{\Delta P(0)}{P_C} \frac{V_g}{V_{\text{gap}}}, \quad (\text{D.12})$$

or the rod is completely filled with water,

$$f = \frac{\Delta P(0)}{\beta}, \quad (\text{D.13})$$

where $f = \gamma t^* / l$ (see Appendix C). Eq. (D.11) is subject to the condition, $\tau \leq l$, or equivalently, $f[1 - \exp\{-t/t^*\}] \leq 1$. Since the exponential term rapidly approaches 0, this condition reduces to $f \leq 1$. In fact, f represents the fraction of the inventory N_{go} available for release during the given time step (see Section 2.2.3). When $f = 1$, all of the gap inventory is released in the time step where Eq. (D.11) reduces to a simple first-order rate model (compare with Eq. (B.4)).

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