

Kinetic simulation of fission product activity in primary coolant of typical PWRs under power perturbations

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Abstract

Kinetic simulations of fission product activity in primary circuits of a typical PWR under power transients, has been performed. A detailed two-stage model-based methodology has been developed and implemented in a computer coder FPCART which uses LEOPARD and ODMUG codes as subroutines. For normal constant power operation, results for over 39 fission products show that the activity due to fission products in fuel region of PWRs is dominated by ^{134}I which is followed by ^{134}Te and ^{133}I . The value of the total fission product activity in fuel region predicted by FPCART code has been found to agree with-in 0.36% range with the corresponding values found by using the ORIGEN-2.0 code. The predictions of FPCART code have also been found in good agreement with the corresponding values found in ANS-18.1 Standard as well as with some available power-plant operation data with 2.4% deviation in the value of specific activity of the dominating fission product ^{134}I . The saturation value of the fission product activity in coolant depends strongly on the fuel-clad gap escape rate coefficient (ε) and approaches a maximum value with increasing value of ε . During power transients, the FPCART predictions have been found in good agreement with the corresponding experimental measurements of ^{131}I specific activity for Beznau and Surry PWRs.

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1. Introduction

Over the past few decades, there has been significant decrease in the steam generator dose rates in various nuclear power plants operating globally. However, the pressurized water reactors (PWRs), which comprise more than two-third majority of operating power reactors worldwide, still maintain 10–100 times higher steam generator dose rates as compared with the competing gas- or sodium-cooled reactors. As a consequence, the maintenance period gets prolonged which not only reduces its effectiveness but also has economic repercussion estimating into several million dollars per such reactor annually (Moore, 1984). The corrosion products, coolant activation and fission product activity in the primary loops of PWRs have been identified as the dominant contributors toward the high dose rates in these systems (Comley, 1985; Lister et al., 1985; Cohen, 1969).

The diffusion of radioactive rare gas fission products from UO_2 fuel and their subsequent leakage from cladding into pri-

mary coolant has been of concern from early reactor operation period (Findlay et al., 1970). In order to gain fundamental understanding of the physical processes involved in fission product release from defective fuel, purposely defective fuel elements were used during an in-reactor research program and as the result, a technique was developed enabling the distinction of surface uranium contamination from the failed fuel pins (Lewis, 1988). In view of the importance of the fission product releases both during normal operation as well as in severe accidents, the Phebus project was launched in 1992 with single aim, to-investigate the phenomenon of fuel damage and fission product release under severe power reactor accident conditions (Hardt and Tattegrain, 1992). The fission product releases have also been observed in research reactors including HEU fueled SLOWPOKE-2 core where transportable gamma-ray spectrometric data was collected and later analysis identified the source of releases as the exposed uranium bearing surfaces (Harnden-Gillis et al., 1994). The current experimental effort is mainly focused on detailed studies of the transport and release of fission products under various conditions in a variety of situations.

In past, a lot of theoretical effort has been made towards release modeling for the development of primary coolant

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radioactivity monitoring systems. For this purpose Koo et al., have developed a model for the release of unstable fission products from defective fuel rods into the coolant of PWRs (Koo et al., 1994). They have used both deterministic as well stochastic approaches in their model. Ivanov has proposed a model for the release of fission gases out of porous fuel and has found close comparison between theoretical and experimental values of the characteristic time for flow changes (Ivanov, 1998). Andrew et al., have proposed an artificial neural network based model for estimating fission product releases during severe accident conditions for both CANDU as well as LWR fuel (Andrews et al., 1999).

Based on these models, computer programs have been developed for the estimation of both the number and the degree of failures in operating reactors. In view of the recent advances in reactor design, fourth order Runge–Kutta algorithm-based methodology has been proposed for solution of chain-model of fission products and to estimate their affect on reactivity (Aboanber, 2001). Significant discrepancies have been found in various theoretical predictions and the corresponding experimental data including clad-rupture time, the degree of oxidation and in the amount of liquefied and relocated fuel (Clément et al., 2003). These discrepancies clearly indicate deficiencies in the models used for predictions. Over the years, much research effort has been devoted towards obtaining information about failed fuel fraction from the primary coolant activity. However, the available computer codes for this purpose contain significant overall errors (Chun et al., 1998). Reliable codes giving estimates of primary coolant activity can clearly be used for on-line monitoring and to schedule a timely replacement of failed fuel rods from reactors.

In this work a two-stage, fourth order Runge–Kutta technique based methodology has been developed for simulation of the transient behavior of fission product activity in the primary coolant of typical PWR under power perturbations. The details of this methodology along with its computer implementation are presented here. The mathematical details of model including the governing differential equations are given in Section 2. The numerical scheme used for computer simulations is described in Section 3. Results of simulation for primary loop of a typical PWR are given in Section 4. Comparison of the results obtained by using this methodology, have been carried out with existing data where available. The conclusions of this study are given in Section 5.

2. Mathematical model

In this work, a typical 300MWe PWR has been considered. The design specifications of such a system are briefly listed in Table 1. The fission product inventory in the fuel has been modeled by extending the standard equations (Chun et al., 1998) as

$$\frac{dN_{F,i}}{dt} = FY_i P + \sum_{j=1}^{i-1} f_{ij} \lambda_j N_{F,j} - (\lambda_i + v_i + \sigma_i \phi) N_{F,i},$$

$$i = 1, 2, \dots, 4, \quad (1)$$

Table 1

The design parameters of a typical pwr system (Glasstone and Sesonske, 1994)

Parameter	Value
Specific power (MW _{th} /kg U)	33
Power density (MW _{th} /m ³)	102
Core height (m)	4.17
Core diameter (m)	3.37
Number of fuel assemblies	194
Rods per assembly	264
Fuel type	UO ₂
Clad type	Zircoloy
Lattice pitch (mm)	12.6
Fuel rod outer diameter (mm)	9.5
Average enrichment (w/o)	3.0
Flow rate (mg/s)	18.3
Linear heat rate (kW/m ²)	17.5
Coolant pressure (MPa)	15.5
Inlet coolant temperature (°C)	293
Outlet coolant temperature (°C)	329

in the gap region:

$$\frac{dN_{G,i}}{dt} = v_i N_{F,i} + \sum_{j=1}^{i-1} f_{ij} \lambda_j N_{G,j} - (\lambda_i + D\varepsilon_i + \sigma_i \phi) N_{G,i},$$

$$i = 1, 2, \dots, 4, \quad (2)$$

and, in the coolant region:

$$\frac{dN_{C,i}}{dt} = D\varepsilon_i N_{G,i} + \sum_{j=1}^{i-1} f_{ij} \lambda_j N_{C,j}$$

$$- \left(\lambda_i + \frac{Q}{W} \eta_i + \beta + \tau \sigma_i \phi + \frac{L}{W} \right) N_{C,i},$$

$$i = 1, 2, \dots, 4, \quad (3)$$

where $N_{F,i}$ is the number of i th radionuclide atoms in fuel (indicated by subscript F), $N_{G,i}$ the number of i th radionuclide atoms in fuel-clad gap (indicated by subscript G), $N_{C,i}$ the number of i th radionuclide atoms in primary coolant (indicated by subscript C), F the average fission rate (fissions/W s), Y_i the Fission yield of the i th radionuclide, P the reactor core thermal power (W), λ_i the Decay constant of the i th radionuclide, v_i the escape rate coefficient for the i th radionuclide, σ_i the microscopic abs. cross section for the i th radionuclide (cm²), $f_{ij} \equiv i \rightarrow j$ Branching ratio, D the failed fuel fraction, τ the (coolant residence time in core)/(total primary circuit time), β the bleed-out fraction for boron control, ϕ the neutron flux (#/cm² s), L the coolant leakage rate (g/s), w the total coolant mass (g), η_i the resin purification efficiency for i th radionuclide and Q is the let-down flow rate (g/s).

The values of these parameters used in simulations have been listed in Tables 2 and 3.

3. Calculational methodology

Based on the above system of ordinary differential Eqs. (1)–(3), a computer program FPCART (Fission-product primary coolant activity in reactor transients) has been developed in this

Table 2
Values of various operational parameters used in these simulations

Parameter	Value
D	2.5×10^{-3}
L (g/s)	2.3
Q (g/s)	470
β	0.001
W (g)	1.072×10^9
V (cm ³)	1.485×10^9
τ	0.056
P_0 (MW _{th})	998
F (fission/W s)	3.03×10^{10}

work. The program is written in Fortran-77 for personal computers. The FPCART program reads job specific data at the initialization stage. It then computes group constants using core design parameters given in Table 1. For this purpose, it uses the LEOPRAD code (Barry, 1963) which is a zero-dimensional unit cell program employing 54 fast and 172 thermal groups. It uses evaluated nuclear data file (ENDF-IV) as cross section library. The cell-averaged group constants are then passed on to the ODMUG code (Thomas and Edlund, 1980) which is also used

as a subroutine in the FPCART code. The ODMUG program solves 1D multi-group diffusion equation and computes spatial profile of group fluxes in the reactor core. Subsequently, these group fluxes are averaged over the core and the resulting values are used as parameters in the transient governing equations for fission product activity.

The FPCART program then solves the large coupled system of ordinary differential Eqs. (1)–(3) using the fourth order Runge–Kutta method to find the activity values due to various fission products/progeny in the fuel, gap and in the primary coolant. These calculations can be performed both for steady state as well as for various transients.

4. Simulation results

The simulation results of the FPCART code presented here are based on a typical 1000 MW_{th} PWR with zero initial fission product activity throughout the system. The design data for a typical PWR are given in Table 1 (Glasstone and Sesonske, 1994). The computer codes LEOPARD (Barry, 1963) and ODMUG (Thomas and Edlund, 1980) were used as a subroutine in the FPCART code for the estimation of core averaged group fluxes.

Table 3
Data for various fission product gases and associated decay products

Isotope (location in chain)	$T_{1/2}^a$	η	γ^b	ν	σ (b) ^b	Branching ratios, f_{ij}				
						1 → 2	1 → 3	2 → 3	2 → 4	3 → 4
Kr-85M (1)	4.48 h	0.4538	1.3E–2	6.5E–8	0.0	1.0	0.0			
Kr-85 (2)	10.752 years	3.958E–5	1.3E–2	6.5E–8	1.84E–1			1.0		
Kr-87 (3)	1.272 h	0.7454	2.56E–2	6.5E–8	5.518E+1	1.0				
Kr-88 (1)	2.84 h	0.5674	3.55E–2	6.5E–8	0.0	1.0	0.0			
Kr-89 (1)	3.15 months	0.986	4.63E–2	6.5E–8	0.0	1.0	0.0			
Rb-89 (2)	15.4 months	0.9	2.05E–3	1.3E–8	0.0			1.0	0.0	
Sr-89 (3)	50.7 days	0.986	1.75E–4	1.0E–11	5.264E–2					1.0
Sr-90 (1)	28.79 years	0.9	7.58E–2	1.0E–11	8.739E–2	1.0	0.0			
Y-90 (2)	2.671 days	0.9	8.97E–8	1.6E–12	5.93E–1			1.0	0.0	
Sb-129 (1)	4.36 h	0.9	5.43E–3	1.0E–11	0.0	0.166	0.834			
Te-129M (2)	33.6 days	0.9	1.4E–7	1.0E–9	2.90E–1			0.63		
Te-129 (3)	1.16 h	0.9	5.11E–3	1.0E–9	0.0				0.37	
I-129 (4)	1.61E7 years	0.99	5.0E–6	1.3E–8	5.225					1.0
Te-131M (1)	1.25 days	0.9	1.1E–2	1.0E–9	0.0	0.21	0.79			
Te-131 (2)	25.0 months	0.9	1.71E–2	1.0E–9	0.0			1.0	0.0	
I-131 (3)	8.023 days	0.99	3.29E–5	1.3E–8	3.229E–1					0.0109
Xe-131M (4)	11.93 days	8.923E–3	4.05E–4	6.5E–8	0.0					
Sb-132M (1)	4.1 months	0.9	1.07E–2	1.0E–11	0.0	0.0	1.0			
Sb-132 (2)	2.8 months	0.9	1.67E–2	1.0E–11	0.0			1.0	0.0	
Te-132 (3)	3.204 days	0.9	1.54E–2	1.0E–9	4.89E–4					1.0
I-132 (4)	2.28 h	0.99	2.06E–4	1.3E–8	0.0					
Te-133M (1)	55.4 months	0.9	3.06E–2	1.0E–9	0.0	1.0	0.0			
I-133 (2)	20.8 h	0.99	4.9E–2	1.3E–8	0.0			0.0285	0.9715	
Xe-133M (3)	2.19 days	4.663E–2	2.67E–5	6.5E–8	0.0					1.0
Xe-133 (4)	5.244 days	2.001E–2	4.9E–02	6.5E–8	2.44E+1					
Te-134 (1)	41.8 months	0.9	6.97E–2	1.0E–9		1.0	0.0			
I-134 (2)	52.8 months	0.99	7.83E–2	1.3E–8				1.0	0.0	
Te-135 (1)	19.0 s	0.9	3.11E–2	1.0E–9	0.0	1.0	0.0			
I-135 (2)	6.57 h	0.9	2.98E–2	1.3E–8	2.119E–3			0.1651	0.8349	
XE-135M (3)	15.29 months	0.9098	1.10E–2	6.5E–8	0.0					
XE-135 (4)	9.14 h	0.2205	6.54E–2	6.5E–8	2.445E+5					0.994

^a Croff (1980).

^b JEFF 3.1 (2005).

The values of various simulation parameters including failed fuel fraction, purification/removal rates, coolant leakage rate etc. have been taken as shown in Table 2.

Thirty-nine different fission products and their progeny have been considered in this study encompassing all dominant isotopes from dose/exposure and radiological consequences perspective. These include various isotopes of iodine, caesium, strontium, xenon, krypton etc. During normal full power operation of nuclear reactor, these isotopes buildup to their saturation values at various times depending on their characteristic half-lives. The saturation value of the quantity of these radioisotopes depends on the flux levels, the corresponding half lives and their fission yield (Tables 3 and 4).

4.1. Fission product inventory in fuel for constant power operation

For an extended full power operation of nuclear reactor, the build-up of fission product inventory in fuel takes place (Fig. 1). The FPCART code simulations for this buildup have been compared with the ORIGEN code (Croff, 1980). The quantities of various radioisotopes at the end of a 200 h constant power run are given in Fig. 2. These results show good agreement between simulation values of the FPCART and the corresponding results obtained by using the ORIGEN code. It is found that ^{137}Cs is at the top of the list and is followed by ^{90}Sr and ^{133}Xe . These are followed by ^{89}Sr , ^{131}I and ^{132}Te .

The FPCART simulated values of activity of various radioisotopes have been compared with the corresponding results of

the ORIGEN code in Fig. 3. Since, apart from quantity of radionuclide, the activity includes decay constant as a factor, therefore, the activity profile shows separate behavior than the corresponding quantity profile. In this case, ^{134}I is found at the top of the list and is followed by ^{134}Te and ^{133}I . The results of FPCART code are generally in good agreement with the corresponding values found with the ORIGEN code. Some deviations are seen in the case of ^{134}Te (~4.8%), ^{137}Xe (~3.6%) and ^{88}Rb and ^{88}Kr (~3.3%) which are mainly due to differences in the corresponding values of the fission yields. The total value of activity calculated by the FPCART code agrees with the corresponding value found by the ORIGEN to within 0.38% indicating high reliability of FPCART code simulations (Fig. 4).

FPCART simulations show saturation behavior of fission product activity in primary coolant. The saturation value depends on the prevailing power level and is expected to change in power transients. The variation of specific activity of ^{132}I in primary coolant with time has been studied with FPCART program. Initially, the reactor was operated at constant power level for 2500 h during which the ^{132}I activity in coolant reached its saturation value. After this time, power transient was introduced. Four power transients were considered for this study including a 15% over-power step and three over-power ramps. In each case, the specific activity values approached a higher saturation value and time taken for this purpose is found to depend on the rate of reactivity insertion. In case of step reactivity insertion, the new higher level is reached in shortest time where as for fast- and slow-ramps, longer time periods are needed.

Table 4
Comparison of the FPCART code predicted saturation value of specific activity of various isotopes in primary coolant of a typical PWR with the ANS-18.1 standard and some experimentally measured data

Isotope	λ (s^{-1})	This work ($\mu\text{Ci/g}$)	Standard ANS-18.1 ($\mu\text{Ci/g}$) ^a	Measured ($\mu\text{Ci/g}$) of coolant				
				Surry unit-1 ^b	Turkey point unit-3 ^c	Turkey point unit-4 ^c	H.B. Rob. unit-2 ^b	R.E. Ginna-Plant ^b
Kr-85M	0.005	1.62E-1	1.6E-1		1.8E-2	1.7E-2	7.94E-3	0.08
Kr-85	0.005	4.58E-2	4.3E-1	2.01E-1		<8.0E-4		
Kr-87	1.0E-3	1.49E-1	1.5E-1	7.86E-4	3.4E-2	2.5E-2	6.74E-3	0.20
Kr-88	4.5E-3	2.74E-1	2.8E-1		4.7E-2	3.8E-2	9.45E-3	0.22
Sr-89	2.5E-6	2.27E-5			1.9E-5	8.9E-5		
Sr-90	1.0E-7	2.65E-7			2.1E-7	3.3E-7		
Te-129M	3.0E-6	2.43E-5			2.7E-5	<3.0E-5		
Te-129	2.5E-3	1.10E-2			1.0E-2	<1.0E-2		
Te-131M	4.0E-5	1.50E-4			1.5E-4	<2.0E-4		
Te-132	7.0E-7	2.93E-5			2.5E-5	3.0E-5		
I-131	6.0E-5	4.21E-2	4.5E-2	8.26E-4	1.1E-2	8.0E-3	2.07E-3	0.07
I-132	4.5E-3	2.08E-1	2.1E-1		1.4E-1	1.9E-2	1.49E-2	0.79
I-133	7.0E-4	1.50E-1	1.4E-1	4.89E-3	8.8E-2	1.7E-2	8.40E-3	0.46
I-134	1.5E-2	3.32E-1	3.4E-1	1.17E-3	2.6E-1	2.2E-2	3.03E-2	1.24
I-135	5.0E-3	2.12E-1	2.6E-1	5.20E-3	1.5E-1	1.9E-2	2.14E-2	0.75
Xe-131M	0.8	3.94E-2	7.3E-1		<2.0E-3	3.0E-4		
Xe-133M	1.5E-3	7.02E-2	7.0E-2		4.3E-3	6.6E-3	3.28E-3	
Xe-133	3.4E-4	2.60	2.60	1.42E-2	2.0E-1	2.1E-1	1.75E-1	1.28
Xe-135M	2.0E-2	1.25E-1	1.3E-1		1.1E-1	2.1E-2		0.12
Xe-135	3.5E-3	8.20E-1	8.5E-1		1.5E-1	1.2E-1	3.82E-2	0.81

NUREG/CR-6261 (1995).

^a ANSI/ANS-18.1-1984.

^b Salvatori, WCAP-8253 (1974).

^c NUREG/CR-1992.

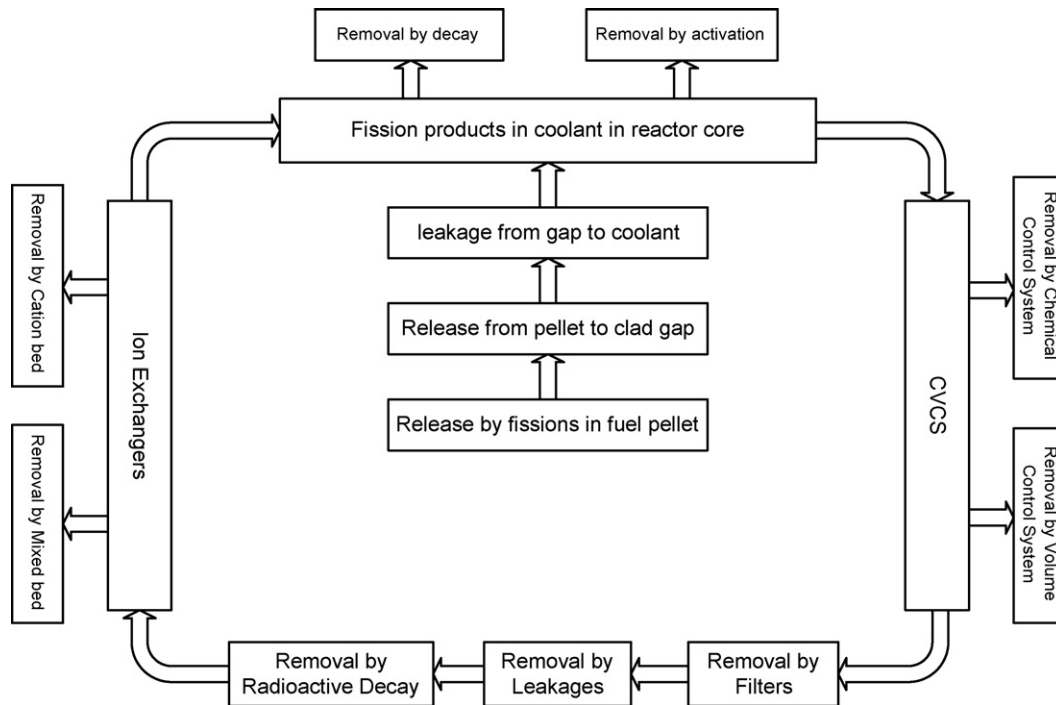


Fig. 1. Schematic diagram of production and loss cycle of fission product activity in primary coolant circuit of PWRs.

In the case of ^{135}Xe , the positive reactivity insertion over-power transient has more complex behavior as shown in Fig. 5. Just after insertion of step reactivity, the values of specific activity show a decreasing behavior which is mainly due to initial domination of removal-rates of ^{135}Xe at higher flux levels. Gradually, the production rates of ^{135}Xe starts rising due to higher production rate of its parent ant its subsequent decay and the corresponding value of ^{135}Xe specific activity approaches a new higher level. After 200 h of operation, the reactor is scrammed and due to absence of its removal by absorption at this stage, the levels of ^{135}Xe are found to rise to a much higher level and fall later due to its natural decay. For slow ramp-insertion, the new higher level of specific activity falls short in comparison with the

step-insertion and as a result, the corresponding post-shutdown peaking also remains lower.

The dependence of the value of saturation value of i th-isotope specific activity in coolant region on the escape rate coefficient (ε_i) from fuel-clad gap has been investigated in this work. After applying the steady state condition on the set of Eqs. (1)–(3), and by elimination of variables, the following analytical expression is found for the saturation value of the specific activity for the i th radionuclide in the coolant ($i = 1, 2, \dots, 4$):

$$N_{C,i}^{\text{sat}} = \frac{D\varepsilon_i N_{G,i}^{\text{sat}} + \sum_{j=1}^{i-1} f_{ij} \lambda_j N_{C,j}^{\text{sat}}}{\lambda_i + Q/W\eta_i + \beta_i + \tau\sigma_i\phi + L/W}, \quad (4)$$

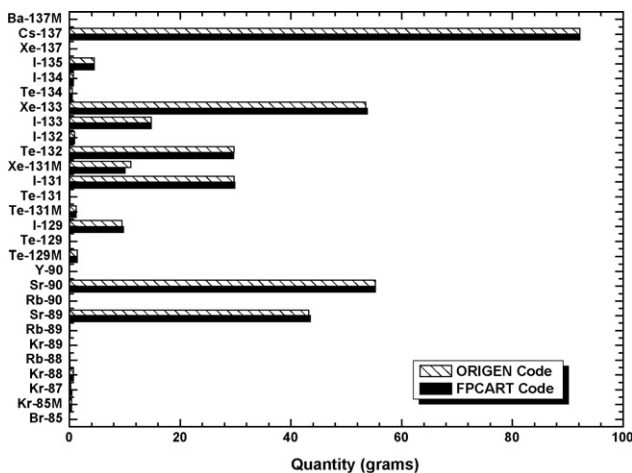


Fig. 2. Comparison of FPCART predicted values of the quantity of various leading fission products in fuel region with the corresponding values obtained by the ORIGEN code.

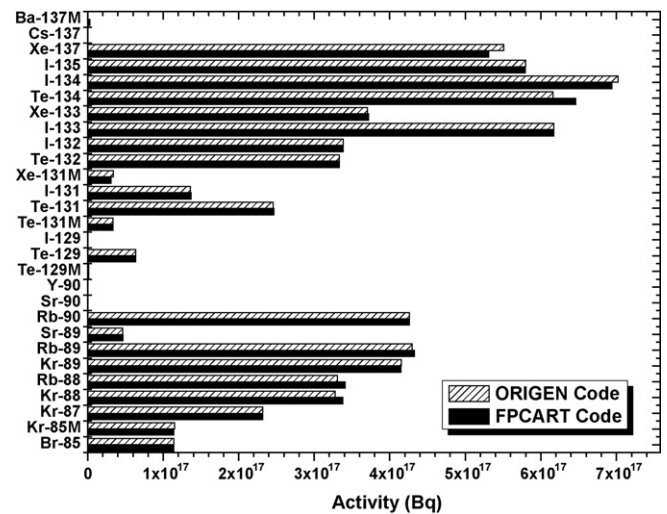


Fig. 3. Comparison of the FPCART computed values of fission product activities of various dominating radionuclides in fuel region with corresponding results from the ORIGEN code.

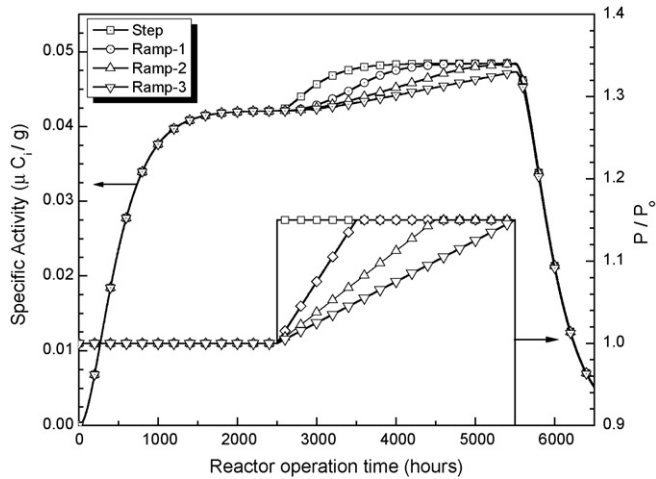


Fig. 4. Variation of the specific activity of ^{132}I with time for various reactor power transients including step and ramps.

where

$$N_{G,i}^{\text{sat}} = \frac{v_i N_{F,i}^{\text{sat}} + \sum_{j=1}^{i-1} f_{ij} \lambda_j N_{G,j}^{\text{sat}}}{\lambda_i + D\varepsilon_i + \sigma_i \phi}, \quad (5)$$

and

$$N_{F,i}^{\text{sat}} = \frac{F Y_i P + \sum_{j=1}^{i-1} f_{ij} \lambda_j N_{F,j}^{\text{sat}}}{\lambda_i + v_i + \sigma_i \phi}. \quad (6)$$

Clearly it has non-linear behavior due to presence of ε_i in both numerators as well as in the denominator. The variation of saturation values of specific activity with ε_i has been studied by using the FPCART program. These results have been compared with the corresponding values found by Eqs. (4)–(6) as shown in Fig. 6. The FPCART predictions are in excellent agreement with the corresponding analytical values throughout the range of ε_i values studied. The saturation values of activity show a linear rise for small values of ε_i and then approaches a maximum limiting value for large values of ε_i . In order to use the predictions of the simulations for estimation of failed fuel fraction and degree of

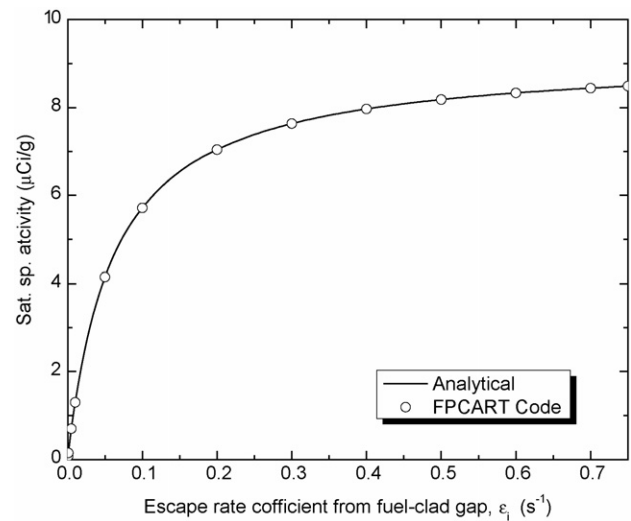


Fig. 6. Comparison of FPCART predicted dependence of saturation values of ^{87}Kr specific activity on escape-rate coefficient ε_i from fuel-clad gap, with the corresponding analytical results.

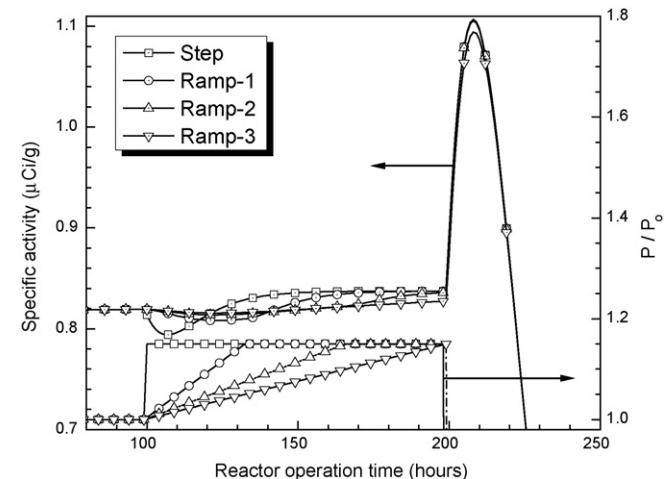


Fig. 5. Time variation of ^{135}Xe specific activity for various reactor power transients including step and ramps.

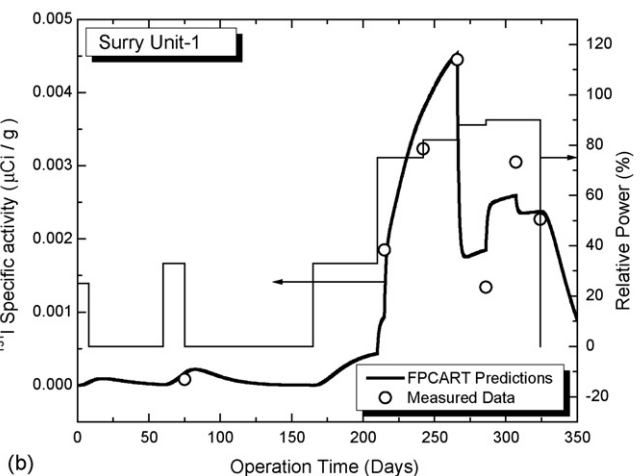
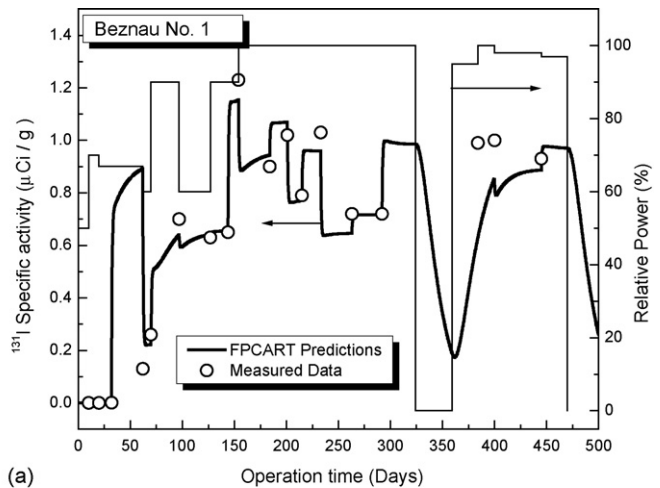


Fig. 7. Comparison of FPCART predicted values of ^{131}I specific activity with the corresponding experimental measurements for: (a) Beznau no. 1 (cycle-1) and (b) Surry no. 1 (cycle-1) units.

failure of fuel, the saturation values of the specific activity should have high sensitivity on ε_i . Also, it is desirable to have the range of high sensitivity for small values of ε_i since one would like to locate and quantify fuel failure well before complete failure of fuel pin. From the results shown in Fig. 6, it is clear that the high sensitivity range is available for small values of ε_i , which is consistent with the requirements.

The prediction of FPCART program were compared with the experimentally measured values of the ^{131}I specific activity in primary coolant circuits of Beznau-1 as well as Surry-1 PWRs for the first cycle in each case (Salvatori, WCAP-8253) during power transients. The corresponding comparison is shown in Fig. 7(a) and (b), respectively, utilizing experimental data based values of 'D', at each time interval. The FPCART predictions are generally in good agreement with the corresponding experimental results. A similar agreement has also been obtained when the FPCART predictions were compared with available experimental data of some other plants as well.

5. Conclusions

- The transient behavior of fission product activity in primary coolant of a typical PWR has been studied in this work. For this purpose, a two-stage methodology has been developed and implemented in a Fortran-77 based computer program FPCART.
- Results for specific activities of over 38 dominant fission products and their progeny studied for normal operating conditions generally show a rapid rise towards saturation values. While ^{134}I , ^{134}Te and ^{133}I are the main contributors of activity in the fuel region, the primary coolant activity has been found to be dominated by ^{133}Xe and ^{135}Xe .
- The predictions of FPCART program have generally been found in good agreement with the corresponding experimental results for ^{131}I specific activity in primary circuits of various operating PWRs during power transients.
- The simulations show strong dependence of primary coolant activity on prevailing power level. For step-as well ramp-overpower transients, the primary coolant specific activity generally shows approach towards a higher saturation level.
- Sensitivity analysis of the saturation values of specific activity due to fission products revealed its strong dependence on the escape rate coefficient from fuel-gap (ε_i), especially towards lower range of values of ε_i . This is highly desirable for the localization and quantification of failed fuel in an operating PWR.

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