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Recent Trends in Mathematical Modeling and Simulation of Fission Product Transport From Fuel to Primary Coolant of PWRs

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1. Introduction

With over 437 operational power plants, nuclear systems contribute 370705 MW(e) worldwide [1]. The Pressurized Water Reactors (PWR) constitute a two-third majority of the operational nuclear power plants while the nuclear reactors in planning and construction phases also show strong trend towards PWRs. These systems are mainly used as baseline load carriers while conventional fossil fueled systems are used for load adjustments and variations [2]. The PWRs have higher than average levels of radiation fields emanating from the corrosion and fission product activity [3] [4] [5]. This leads to prolongation of maintenance schedules entailing loss of revenues mounting to several million dollars per plant annually [6]. Consequently, the plant availability factors are also lowered. This situation is further aggravated due to strong shift of plant age profile toward over 25 years operational range. With plant aging, the fuel failures become more frequent which leads to enhancement of radiation levels in the primary circuits of PWRs.

The levels of fission product activity (FPA) have been of concern both from the operational as well as from accidental perspectives. These levels are continuously monitored during the normal operation of PWRs. The fuel pins develop leakages with their burnup. When the failed fuel fraction exceeds a safe limit, replacement of defective assemblies by refueling becomes necessary. Therefore, low levels of leaked-out fission products (FPs) in primary coolant of PWRs are indicative of the core health [7]. In the accidental conditions, the total value of FPA serves as the available source term that potentially can escape into the surroundings [8] [9].

The fission products are released in the fuel matrix during burnup. They escape from the ceramic pellets into the gap between pellets and the clad regions. Hyun et al. [10] have developed an analytic method for the fuel rod gap inventory of unstable fission products during steady state operation of PWRs. The fission gas bubbles escape from grain corners and are interlinked in the open space. The release rate depends on the bubble interlinkage along with temperature and burnup. A generalized model for fission product transport in the fuel-to-sheath gap was given by Lewis [11]. Barrachin et al. presented a review of fission product behaviour in UO_2 fuel [12].

The release of fission products from fuel-clad gap into primary coolant involves clad failure. A model describing pallet oxidation, subsequent enhancement of diffusivity and bubble formation at grain boundaries, their interlinkage and release into open surfaces, was developed by Koo et al [13]. This model is stochastic in nature and incorporates inherent randomness of the underlying physical phenomenon using Monte Carlo method. While the prediction based on this model are in good agreement with the corresponding experimental measurements in the linear heating regime, strong under-predictions have been reported for the remaining regime. The Ivanov's model [14] gave good description of various processes involved in the release of FPs from the porous ceramic fuel, its leakage from clad and mixing with the primary coolant. Theoretical predictions based on this model have been reported in good agreement with the corresponding experimental data.

Combined failures based model has been developed by Clink and Freeburn [15] which was employed in an on-line coolant activity monitoring system. Such systems carryout estimation of failed fuel fractions in non-destructive manner. Normally, these systems are designed for constant power, steady state operational conditions. The Clinck and Freeburn model was observed to under-predict failed fuel fractions even for steady-state operation [16].

A theoretical model has been developed by Tucker and white [17] for the estimation of FPs from ceramic UO_2 fuel. In this model, first, the probabilities of leakages of FPs from fuel interior through grain-edge tunnel pore to outer portions are figured out. These probabilities strongly depend on the interconnectivity of pores in the ceramic fuel. A good agreement has been reported between theoretical predictions made by using this model and the corresponding experimental measurements.

2. FPA simulation codes

In view of the importance of the FPA for normal operation as well as for accidental scenarios, various computer programs have been developed for its estimation. They fall into two basic categories:

- Point depletion codes
- Fission Product Transport Codes
    - Empirical
    - Semi-Empirical
    - Mechanistic

The point depletion codes carryout production, buildup, decay and depletion calculations for a wide variety of radionuclides in the core region. As such, they provide reliable estimates of radioisotope inventory in the reactor fuel. They typically ignore spatial details while retaining spectral details of the neutron field. The widely used WIMS computer code [18] for 1-D transport theory macroscopic group constant generation employs 69-group library along with DSN or Stochastic methodology. It performs details buildup, depletion and burnup calculations for 35 distinct fission products along with one pseudo, lumped fission product. The WIMS code does not perform any further radionuclide transport calculations. The CASMO-4 [19] and DRWIN [20] also belong to the same pin/cell based macroscopic group constant generation codes as WIMS and as far as fission products are concerned, they are limited to radionuclide inventory calculations for the fuel region.

The ORIGEN2 computer code [21] provides extensive radionuclide inventory calculations for 950 fission products along with 120 actinides in point-wise buildup and depletion manner. While one can manually remove or add radionuclides in refueling options, no

attempt is made in the code for the radionuclide transport calculations. An evolved version called MONTEBURNS [22] incorporates spatial details in the depletion/buildup calculations by coupling the ORIGEN2 code with the multipurpose radiation transport code MCNP [23].

The radionuclide transport code category is comprised of three types of computer codes: empirical, semi-empirical and mechanistic codes. In the empirical codes, various data fitting techniques are used for development of empirical models from detailed experimental observations. One advantage of this strategy is that no prior knowledge is required regarding the details of the underlying physical processes involved. At the same time, it gives most accurate results in the sense that they match the experimental results. Consequently, they are extensively used in risk assessment and safety analysis. Lumping of parameters and grouping of similar elements simplifies many features of these codes and adds to their computational efficiency. The MELCOR [24] and CORSOR [25] codes belong to the empirical radionuclide transport class of computer programs. While being highly efficient and reliable, the empirical codes are valid only in a limited range of parameters.

The limitations of the empirical models are relaxed somewhat by incorporating detailed modeling for a part of the simulation while the remaining part is attempted by using empirical approach. The FIPREM [26] computer code attempts fission product transport problem by using empirical Booth equivalent sphere model while detailed diffusion theory based finite difference model is employed for fission product transport into gap region.

The VICTORIA [27] and ECART [28] computer codes, being mechanistic in nature, do not face strict limits of validation. They carryout simulation of radionuclide transport in much broader range of accidental scenarios starting from releases, to dispersion and subsequent deposition. Since these computer programs were specifically designed for accident analysis, therefore, they cannot be used in normal steady-state or in transient cases.

Most of the available computer programs for transport analysis of fission product activity are focused on accidental analysis. For the analysis of fission product transport in the steady state and in transient analysis FPCART-ST computer code has been developed. The details regarding the mathematical modeling, computer implementation and results of simulations carried out using this code are provided here.

3. Kinetic modeling

In these work, a 300 MW(e) PWR has been considered with design specifications as provided in Table 1. The primary circuit of a typical PWR with various indicated essential components is shown in Fig. 1. The reactor is taken with zero levels of FPA in the primary circuit at the start ($t = 0$). The FPA levels in Fuel/Gap/Coolant=F/G/C is governed by the following set of ODEs:

For the fuel region:

$$\frac{dN_{F,i}}{dt} = FY_iP + \sum_{j-1}^{i-1} f_{ij}\lambda_j N_{F,j} - \left[\lambda_i + v_i + \sigma_i\phi\right]N_{F,i} \, , \tag{1}$$

for the gap region:

$$\frac{dN_{G,i}}{dt} = v_i N_{F,i} + \sum_{j-1}^{i-1} f_{ij}\lambda_j N_{G,j} - \left[\lambda_i + D\epsilon_i + \sigma_i\phi\right]N_{G,i} \tag{2}$$

and, for the coolant region:

$$\frac{dN_{C,i}}{dt} = D\epsilon_i N_{G,i} + \sum_{j-1}^{i-1} f_{ij}\lambda_j N_{C,j} + \left[\lambda_i + Q\frac{\eta_i}{W} + \beta + \tau\sigma_i\phi + \frac{L}{W}\right]N_{C,i}. \tag{3}$$

where, 'i' indicates the isotope in the decay chain consisting of four isotopes: $i = 1, 2, ..., 4$. The values of various parameters used in these simulations are listed in Table 2.

In order to compute the saturation values of various radioisotopes in the fuel, gap and coolant regions one can use the following analytical results:

For coolant region:

$$N_{C,i}^{sat} = \left[D\epsilon_i N_{G,i}^{sat} + \sum_{j-1}^{i-1} f_{ij}\lambda_j N_{C,j}^{sat}\right] \bigg/ \left[\lambda_i + \frac{Q}{m}\eta_i + \beta_i + \tau\sigma_i\phi + \frac{L}{m}\right], \tag{4}$$

For gap region,

$$N_{G,i}^{sat} = \left[v_i N_{F,j}^{sat} + \sum_{j-1}^{i-1} f_{ij}\lambda_j N_{G,j}^{sat}\right] \bigg/ \left[\lambda_i + D\epsilon_i + \sigma_i\phi\right], \tag{5}$$

and for fuel region:

$$N_{F,i}^{sat} = \left[FY_i P + \sum_{j-1}^{i-1} f_{ij}\lambda_j N_{F,j}^{sat}\right] \bigg/ \left[\lambda_i + v_i + \sigma_i\phi\right]. \tag{6}$$

Parameter	Value
Specific power (MWth/Kg. U)	33
Number of fuel assemblies	194
In-let coolant temperature (°C)	293
Out-let coolant temperature (°C)	329
Power density (MWth/m³)	102
Fuel pins (rods) per assembly	264
Fuel material	UO_2
Clad material	Zilcoloy
Lattice pitch (mm)	12.6
Fuel pin outer diameter (mm)	9.5
Coolant pressure (MPa)	15.5
Coolant flow rate (Mg/s)	18.3
Linear heat rate (kW/m³)	17.5
Average enrichment (%)	3.0
Core height (m)	4.17
Core diameter (m)	3.37

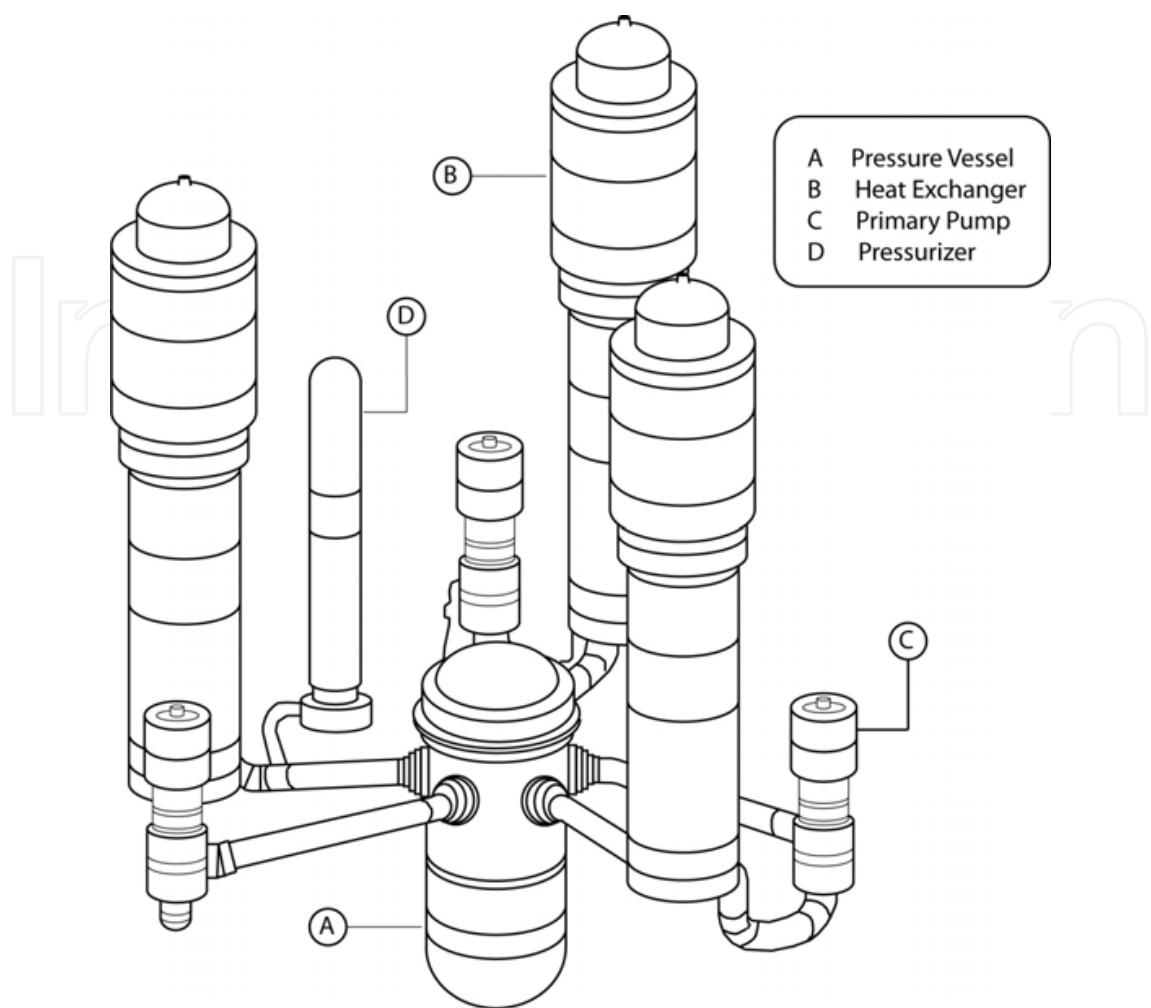Table 1. Design data of a typical pressurized water reactor [37]

Fig. 1. A three dimensional perspective view of a typical PWR primary system with the pressure vessel, heat exchanger, primary pump and pressurizer indicated

Parameter	Value
V (cm^3)	1.485×10^9
L (g/s)	2.3
Q (g/s)	470
D	2.5×10^{-3}
W (g)	1.072×10^9
β	0.001
F $(Fissions/W.s)$	3.03×10^{10}
P_o (MW_{th})	998
τ	0.056

Table 2. Values of different operational parameters used in simulations [37]

3.1 Deterministic computational methodology

Various step involved in the transport of fission products, starting from their release in the fuel matrix, their transport from ceramic pores into the fuel-clad gap, their leakage from clad into the primary coolant, and subsequent removal by leakages, by filters, by radioactive decay etc., is depicted in Fig. 2. The FPA transport model has been implemented in the computer program FPCART. It uses LEOPARD [29] and ODMUG [30] programs as subroutines. The cell averaged multigroup group constant generation is carried out by the LEOPARD subroutine while the group fluxes are found by solution of one-dimensional diffusion equation in the ODMUG subroutine. In the FPCART code, the system of governing ODEs: Eqs. (1) upto (3) are solved numerically using Runge-Kutta (RK) method in this program. The RK-numerical provides efficient time domain solution yielding static as well as dynamic values of FPAs corresponding to about 50 different dominant fission products.
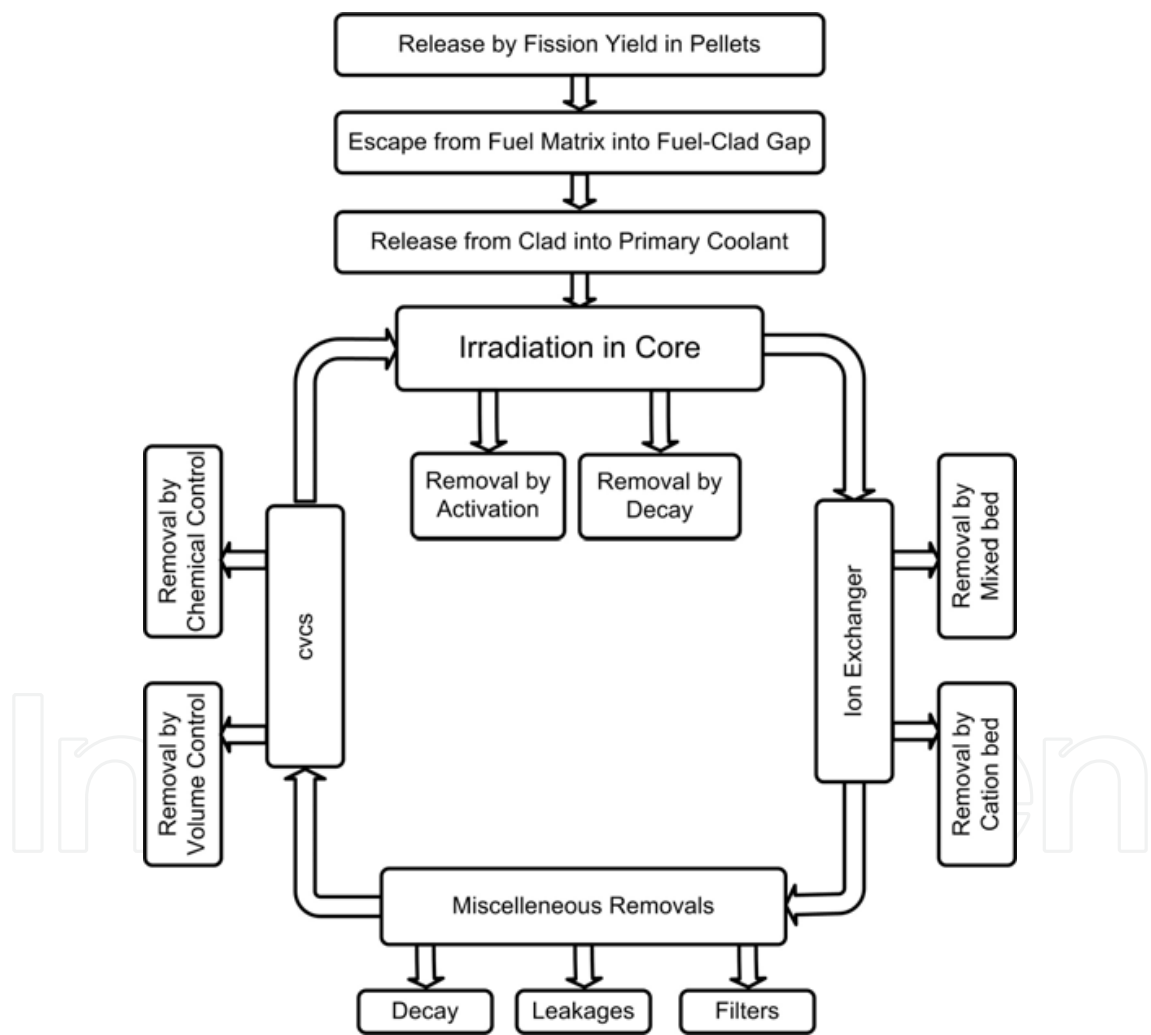

Fig. 2. Block diagram of the fission products production and removal mechanism in the primary circuit of a typical PWR

The computational cycle starts with initialization of the variables with $t = 0$. The group constants are generated by the LEOPARD while the group flux are found using ODMUG. The values of FPAs in the fuel, gap and in primary coolant are initialized as zeros for the

cold clean core. In the time loop, the values of FPAs for about 50 different radionuclides are calculated using RK-scheme for each next time step. The results are stored in separate data files for each fission product chain and for each region. The program allows performing these calculations for power as well as flow rate perturbations.

3.2 Power perturbation model

The FPCART computer code has built-in model for linear power perturbations. This model uses a rate parameter α representing the time rate of change of reactor power. Then, for a time range $[t_{in}, t_m]$ the reactor power is calculated using:

$$P(t) = P_0 f(t) \tag{7}$$

where,

$$f(t) = \begin{cases} 1, & t \leq t_{in} \\ 1 - \alpha[t_m - t], & t_{in} \leq t \leq t_m \\ w_2/w_0 & t > t_m \end{cases} \tag{8}$$

Where
α is slope of the linear change of reactor power;
t_{in} is start of reactor power perturbation;
t_m is end of the reactor power.

3.3 Flow-rate perturbation model

The flow rate perturbation involves primary pump modeling where the balance of angular momentum with the frictional deceleration yields [31] :

$$l\rho \frac{dv}{dt} = -\frac{1}{2} C_f \rho v^2 , \tag{9}$$

where, l is the total length of the loop; ρ is the fluid density; C_f represents total pressure loss coefficient; and v is the fluid speed. The Eq. (9) yields the corresponding solution as flow rate $w(t)$ is:

$$w(t) = w_0 / \left[1 - t/t_p \right], \tag{10}$$

where, w_0 represents the steady state value of flow rate; and $t_p = 2l/(C_f v_0)$ which is typically around 2000 h for transients without boiling crisis.

4. Stochastic release model

The release of fission products from fuel pins is essentially a random process as the time of clad failure, the amount of release as well as the duration of fission product release cannot be specified exactly beforehand. In order to model these aspects in more realistic manner, Monte Carlo based stochastic approach has been used in these simulations. The modified version FPCART-ST is primarily deterministic-stochastic hybrid code. The sampling of fuel pin failure probability distribution function $g(t)$ yields the fuel pin failure time sequence. The intensity function $\psi(t)$ is correspondingly:

$$\psi(t) = g(t)/G(t),\tag{14}$$

where, the cumulative probability distribution $G(t)$:

$$G(t) = \exp\left(-\int_0^t g(s)ds\right),\tag{15}$$

serves the normalization. According to the standard rejection technique [??] the probability of accepting a fuel failure at t_k after t_j is found by using a random number 'η' and comparing it with the ratio 'q':

$$q = \frac{g(t_k)}{g(t_j)},\tag{16}$$

and, if $\eta < q$, this step is repeated otherwise, t_k is accepted as a fuel failure event time. The fuel matrix to gap escape rate coefficient takes the form:

$$\epsilon = D_0\epsilon_0 \exp[-\xi(t - t_0)] + D_F\epsilon_0\tag{17}$$

where, ϵ_0 is the starting value of burst release rate from a punctured fuel rod; ξ represents the characteristic decay constant for the escape rate ; t_0 is time at which the fuel rod fuel rod failure starts; D_F represents the current number of failed fuel rods while $D_0 = 1$ is flag for the failure of the current fuel rod. Typical values of these parameters are: $\xi = 7.2 \times 10^{-5} s^{-1}$ $\epsilon_0 = 10^{-8} s^{-1}$; $D = 60$

5. Results and discussion

5.1 Buildup of fission products in steady-state

The FPCART computer code has been used for the simulation of fission product buildup to steady state saturation values starting with a cold clean core. For a 300 MW(e) typical PWR, the predictions of the FPCART program have been compared with the widely used ORIGEN2 computer code and excellent agreement between the corresponding values has been found. The observed small difference, of the order of a few percent only, can be attributed to difference in the yield of the fission products. The results are shown in Fig. 3. The results indicate dominance of ^{131}I, ^{134}Te, ^{133}I and ^{135}I in the saturation values of fission product activity in the fuel matrix.

5.2 ^{135}Xe activity under step and ramp power transients

With largest absorption cross section, ^{135}Xe acts as dominant poison in nuclear reactors. At the start of operation, the ^{135}Xe levels are zero which climb to saturation levels with time which depend on the power level and time behavior of reactor power during this period. FPCART simulations have been carried out for the study of ^{135}Xe transients for step and ramp power transients. The results are shown in Fig. 4. The ramp power transients lead to somewhat slower rise to saturation levels as compared with the step power changes. For post-scram time periods, the ^{135}Xe levels rise to maximum values; which is followed by gradual decrease.
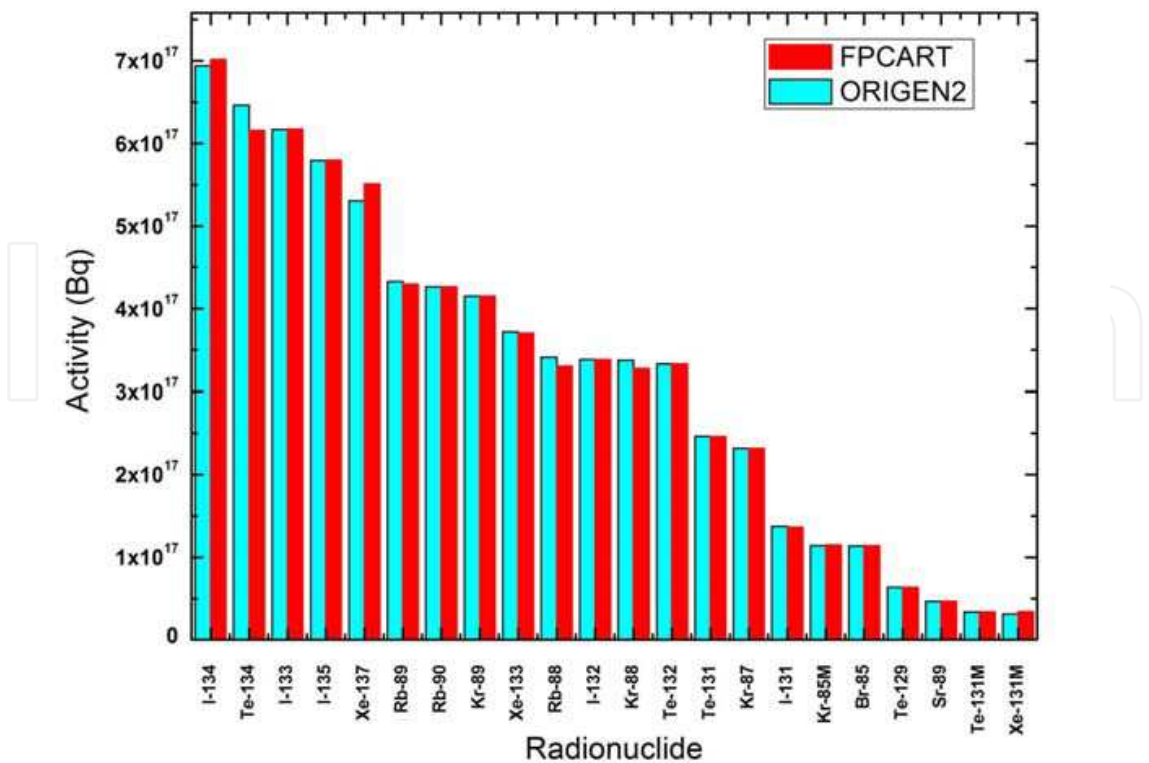
Fig. 3. For steady state operation, FPCART predicted saturation values of activities of various isotopes in PWR fuel with the corresponding computed data using the ORIGEN2 code [35]
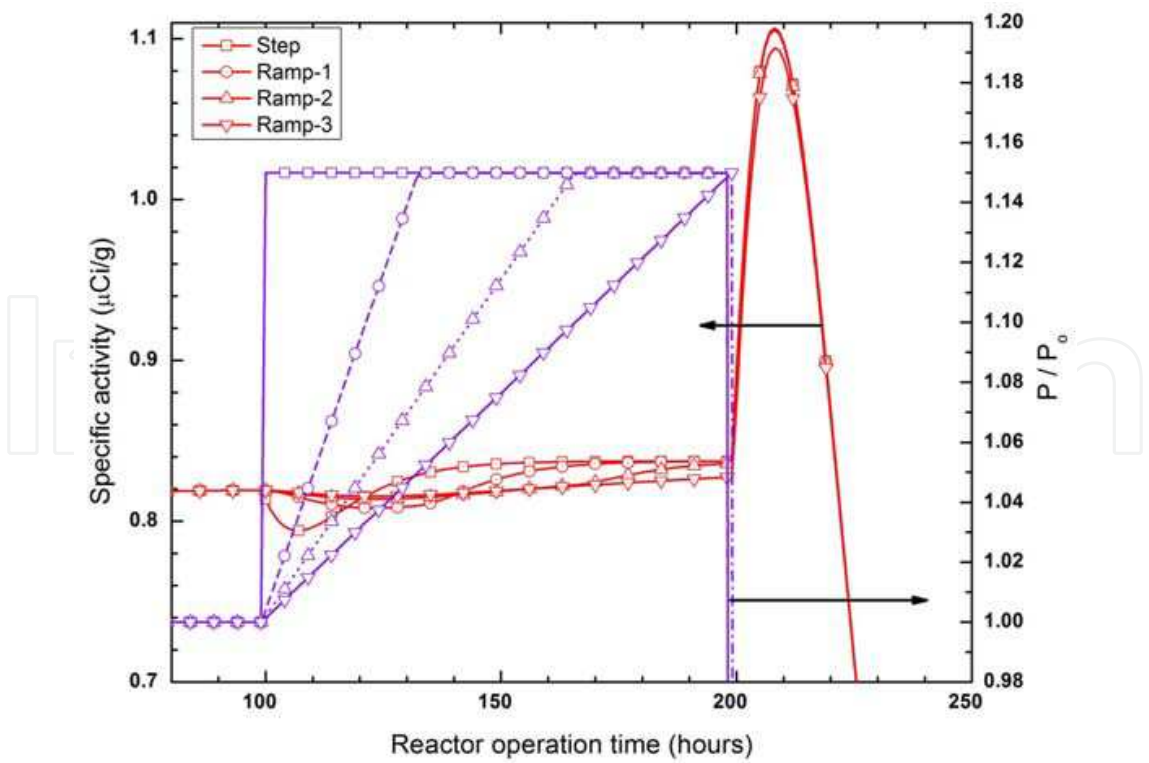

Fig. 4. FPCART simulated variation of ^{135}Xe specific activity with time for step and ramp power transients

5.3 Fission product activity under pump coast-down

The pump-coast down belongs to general class of flow rate transients. During these transients, the core residence time, and total circuit time along with the effective neutron flux values are influenced by the change in flow rate. A decrease in flow rate leads to increase in the fission product activity values. In this study, fifteen different radionuclides belonging to fission products and their decay chains were selected and their approach towards saturation levels was studied under constant power. The pump coast-down was initiated when the levels reached sufficiently close to saturation levels. The corresponding results are shown in Fig. 5 where the isotope-wise as well as total activity variations are shown after the pump coast-down. It is observed that ^{133}Xe is the main contributor having over 40% of total activity. This is followed by [135]Xe, [131M]Xe and [129]Te contributing 12.9%, 11% and 8.2% of the total activity respectively. During the pump coast-down period, the total activity level raises well over 8.6% level before the loss-of-flow signals the reactor shutdown.
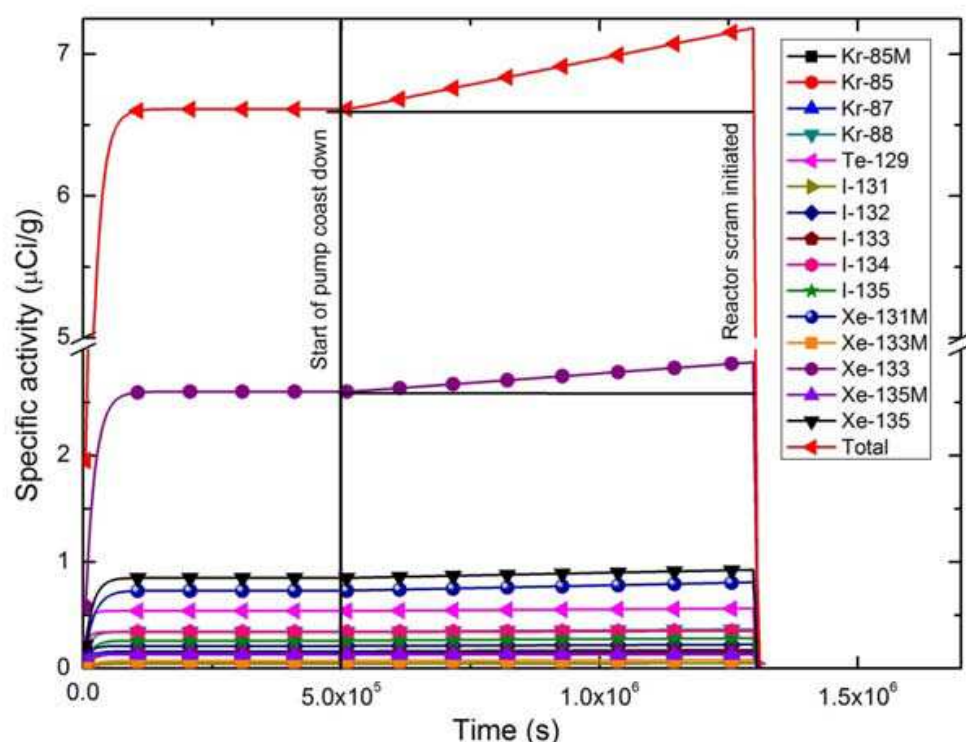

Fig. 5. FPCART simulated primary coolant total activity due to fission products of a 1000 MWth PWR for a $t_p = 2000h$ pump coast-down flow rate transient [36]

5.4 FPCART simulations of FPA under power transients

For validation of the three stage deterministic computational methodology of the FPCART computer code, its predictions were compared against actual experimental data. In the case of BEZNAU (Unit 1) [32] , the FPCART computed time variation of ^{131}I for various power variations during the first cycle have been compared with the corresponding experimental measurements. It is clear from Fig. 6 that FPCART predictions are in good agreement with the experimental data throughout time range. A similar trend has been observed in the case of ^{131}I activity in the ZORITA [32] power plant where again the FPCART predictions have been found in good agreement with the corresponding experimental measurements as shown in Fig. 7.
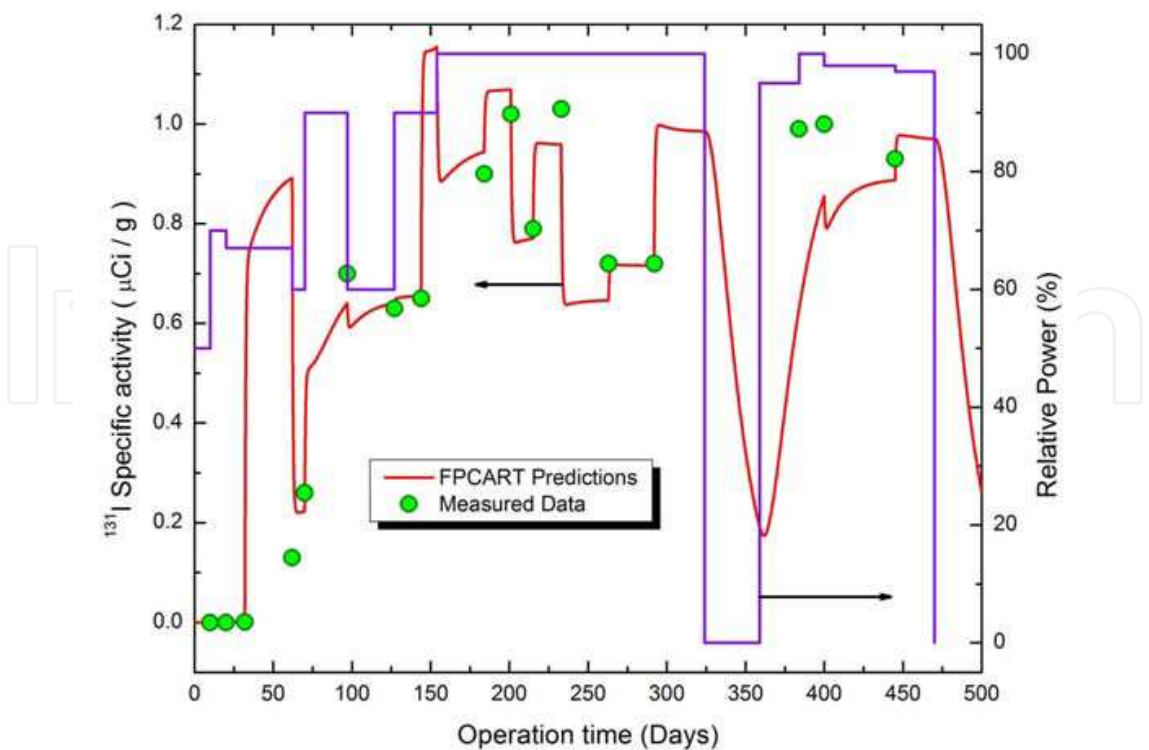
Fig. 6. For power transients, FPCART predicted values of 131I specific activity variations with time compared with the corresponding experimental data for the BEZNAU (Unit-1) power plant

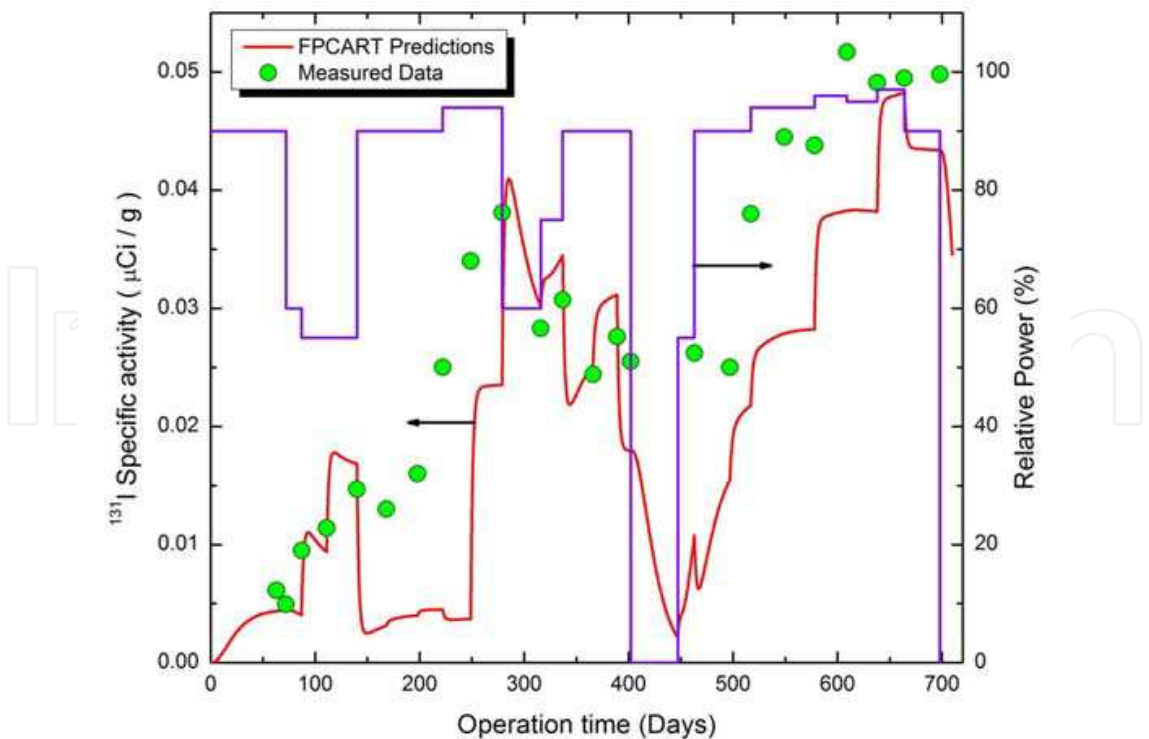Fig. 7. For power transients, FPCART predicted values of ^{131}I specific activity variations with time compared with the corresponding experimental data for the ZORITA power plant

5.5 Stochastic simulations of FPA

A typical PWR has large number of fuel pins arranged in fuel bundles. They can fail at any time due to a wide variety of causes/ reasons. Their failure is essentially a random phenomenon. Therefore, stochastic techniques are well suited for the failure modeling. In this work, a time sequence of their failure has been generated by sampling time dependent intensity function. The escape of fission product and their siblings have been modeled using three step deterministic model of FPCART.
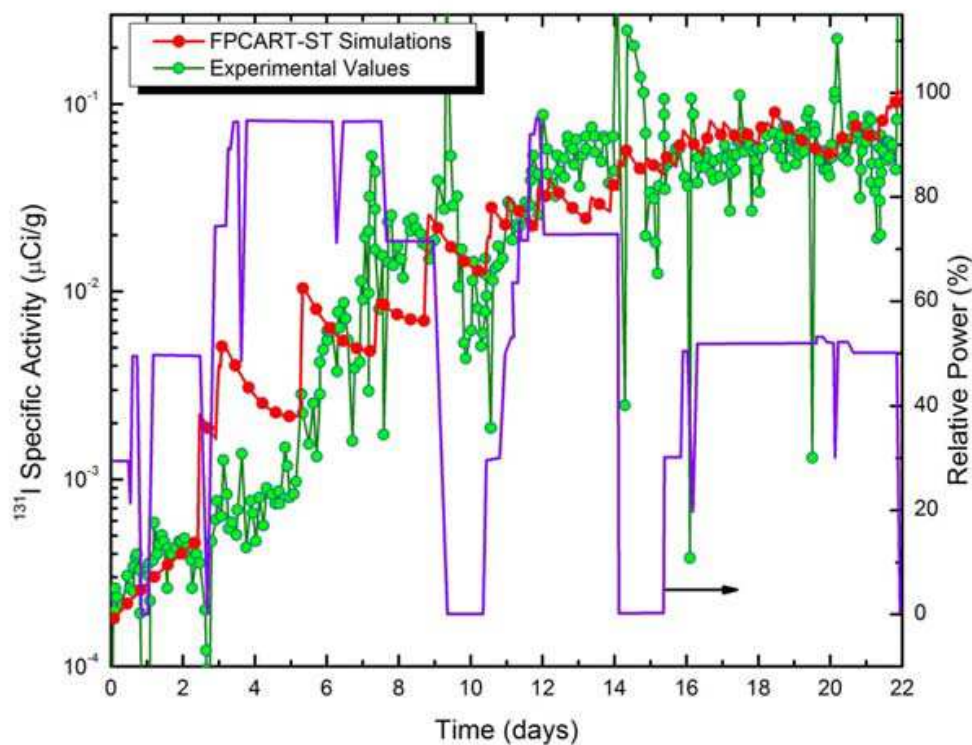

Fig. 8. For power transients, FPCART predicted values of ^{131}I specific activity variations with time compared with the corresponding experimental data for the ANGRA-1 power plant [37]

The Westinghouse designed ANGRA-1 [33] (657 MWe) nuclear power plant was shut-down prematurely in the 4th cycle due to abnormally high levels in the primary coolant. As suspected, one sixth of its core had failed resulting in leakage of fission products and their daughters into the primary coolant stream. The first 22 days of this event have been simulated using the stochastic FPCART-ST computer code. For ^{131}I activity, the predicted values for 70% power levels remain within 15% from the corresponding experimental measurements. Keeping in view the complex nature of the event being simulated, the predictions show good agreement. For larger variations in power level, deviations are found in the predictions which may be attributed to variations in the flow rate that have not been included in this model. The ^{131}I activity spikes found in experimental data remain smeared in the predicted data requiring further investigation regarding couples flow-rate & power transients.

The EDITHMOX [34] experiments, given in the OECD/NEA/IAEA IFPE database, were considered for validation of the FPCART-ST computer code. These experiments are part of a broad experimental program conducted a the France's Siloe research reactor. The EDITHMOX experiment was conducted at the Jet Pompe water loop. Release rates of

various fission products were studied for mixed-oxide fuel under various conditions of burnup and operating power. The FPCART-ST predicted time variation of ^{85}Kr activity for various power levels has been found in good agreement with the corresponding experimental data available from the EDITHMOX experiment. As shown in Fig. 9, the agreement prevails over most of the measured time while some deviations are observed for the first large power drop. This may be attributed to coupling of power transients with flow-rate perturbations which has been ignored in these simulations.
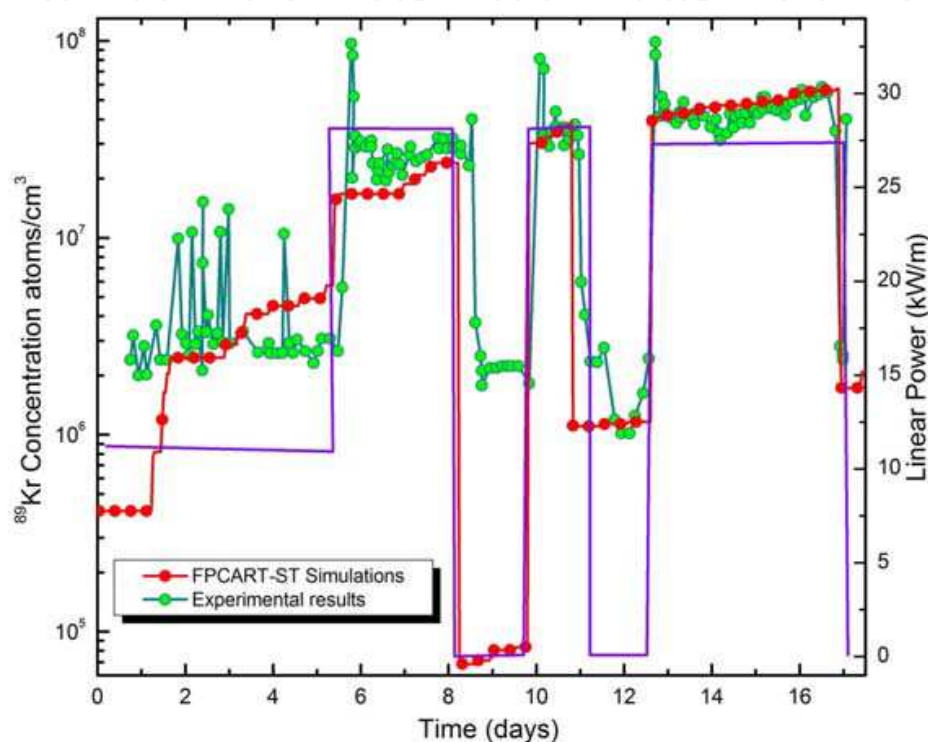

Fig. 9. For power transients, FPCART predicted values of ^{89}Kr specific activity variations with time compared with the corresponding experimental data for the EDITHMOX-1 (Unit-1) power plant [37]

6. Conclusions

The normal operation as well as during the accidental situations, the fission products and their daughters play a dominant role toward potentially imparting high levels of exposure to radiation workers and general public. A review of research effort devoted to modeling and simulation of fission product activity in the primary circuits of typical PWRs has been presented in this work. While the mechanistic models have been found superior in the context of the range of applicability against the empirical and semi-empirical models, the available computer codes are limited to accidental release modeling generally.

Development of methodology for estimation of FPA levels in the primary loops of PWRs during normal steady-state as well as transient conditions has been carried out in this work. The aim of this effort is to model FPA releases into primary coolant in the steady state, as well as in power and flow-rate transients. For this purpose multi-step model has been presented in this work that tracks fission product transport from fuel to fuel-clad gap and finally to the primary coolant. The influence of filters, ion-exchanges, leakages, decay etc.

has been incorporated in this model. For randomly failing fuel pins, stochastic modeling has been carried out for burst releases. The coupled deterministic-stochastic hybrid approach has been found effective for large scale fuel failure events.

The trends towards core life-time extensions and high burn-up cores, coupled with aging of the existing fleet of nuclear reactors; it has become imperative to limit fission product activity to much lower levels. This is required in order to keep PWRs economically feasible against their competitors.

7. Nomenclature

Symbol	Represents
$N_{X,i}$	Number of ith radionuclide atoms in the region: Fuel (X=F), Gap (X=G) and Coolant (X=C)
ϕ	Neutron flux (#/cm².s)
η_i	Resin purification efficiency for the ith radionuclide
Q	Let-down flow rate (g/s)
v_i	Escape rate coefficient of the ith radionuclide
σ_i	Microscopic absorption cross section for the ith radionuclide
Y_i	Fission yield of the ith radionuclide
f_{ij}	Branching ratio $(i \rightarrow j)$ in the ith chain
β	Bleed-out fraction of the primary coolant for boron chemical control
L	The coolant leakage rate (g/s)
W	The total primary coolant mass (g)
λ_i	Decay constant of the ith radionuclide (s^{-1})
F	Average fission rate (fissions/W.s)
P	Thermal power of reactor (W)
D	Failed fuel fraction (#)
τ	Core-to-circuit primary coolant resident time ratio (#)
D_F	Number of failed fuel rods
ξ	Characteristic decay constant for the escape rate (s^{-1})

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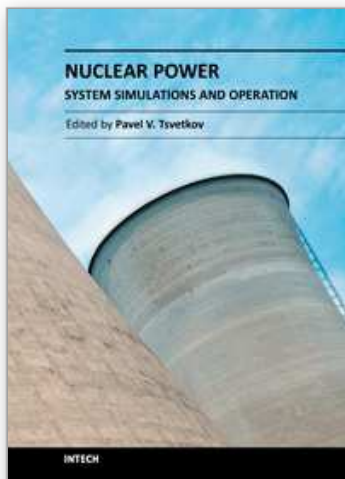
Nuclear Power - System Simulations and Operation
Edited by Dr. Pavel Tsvetkov

At the onset of the 21st century, we are searching for reliable and sustainable energy sources that have a potential to support growing economies developing at accelerated growth rates, technology advances improving quality of life and becoming available to larger and larger populations. The quest for robust sustainable energy supplies meeting the above constraints leads us to the nuclear power technology. Today's nuclear reactors are safe and highly efficient energy systems that offer electricity and a multitude of co-generation energy products ranging from potable water to heat for industrial applications. Catastrophic earthquake and tsunami events in Japan resulted in the nuclear accident that forced us to rethink our approach to nuclear safety, requirements and facilitated growing interests in designs, which can withstand natural disasters and avoid catastrophic consequences. This book is one in a series of books on nuclear power published by InTech. It consists of ten chapters on system simulations and operational aspects. Our book does not aim at a complete coverage or a broad range. Instead, the included chapters shine light at existing challenges, solutions and approaches. Authors hope to share ideas and findings so that new ideas and directions can potentially be developed focusing on operational characteristics of nuclear power plants. The consistent thread throughout all chapters is the "system-thinking" approach synthesizing provided information and ideas. The book targets everyone with interests in system simulations and nuclear power operational aspects as its potential readership groups - students, researchers and practitioners.

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