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Evaluation of Corrosion Product Activity in a Typical PWR with Extended Cycles and Flow Rate Perturbations

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Abstract

Effect of flow rate perturbations has been studied using the modified computer program CPAIR-P for time dependent corrosion rates in Pressurized Water Reactors (PWRs) having extended cycles. In these simulations, a decrease in the corresponding saturation values of corrosion product activity (CPA) is observed for higher pH values. Comparison of CPA's behavior has been done for constant flow-rate case as well as for transients with elevated ^{10}B levels (~40%) in dissolved boric acid in coolant in two operating cycles. When the flow rate is decreased in the first cycle, the saturation value of CPA attains new higher values. Also, in the second operating cycle, the saturation values are about 12% higher when compared with the values in the first cycle.

1. Introduction

Recent Pressurized Water Reactors (PWRs) are aiming at better performance of nuclear plants by increasing the fuel cycle length as high burn-up leads to better utilization of fuel as well as generation of higher revenues [1]. Recent studies indicate that enriched boric acid (EBA) decreases the requirement of high concentrations and the high value of pH (~7.4) can be done with 2.2 ppm of lithium. It also does not introduce the corrosion cracking problems in Pressurized Water Reactors (PWRs) [2] [3] [4]. This can improve the discharged burnup from average value of 33 MWd/kgU to 40 - 50 MWd/kgU.

However, extended cycles put higher safety limits on fuel integrity for longer fuel residence time in core and corrosion induced cracking is one of the major reasons for material failure in PWRs [5]. When the corrosion elements come from the magnetite (Fe_3O_4), the pH levels of 7.3 - 7.4 are needed instead of 6.9. Also, high values of temperature in the system can cause highly corrosive coolant and decomposition of water further increases its corrosion [1] [4]. The corrosion rate in PWRs increases with the effective full power days [6]. The observed increase involves many factors including phase transformations that occur due to aging and decomposition of stainless steel [7] etc. The corrosion elements get activated by circulation in reactor core. Large value of corrosion product activity (CPA) in the primary loop does become a problem to the reactor maintenance [6] [8].

In PWRs, the CPA mainly comes from short-lived ^{56}Mn and ^{24}Na. However; iron, molybdenum and cobalt make almost all of the activity after shutdown of reactor. Table 1 shows some properties of these isotopes [9]. Primary coolant in PWRs maintains very high dose due to the CPA even after the reactor shutdown. Also, the end of cycle coolant activity can further increase due to any transient condition during reactor operation [8] [9]. The soluble forms of ^{60}Co, ^{58}Co and ^{54}Mn show dependence on flow rate, pH values and boron [10] [11].

Several studies have been carried out on coolant activation in nuclear reactors with focus on the flow rate & power transients. Reactor transient studies indicate that reactivity as well as flow rate perturbations result in neutron flux peaking [12] For PWRs, Venz and Weidmann showed that the coolant chemistry is important for extended fuel cycles. Coolant Activity at Beznau nuclear power plant was reduced when the pH levels in the coolant were raised to 7.4 [13].

Table 1. Activation products and their reaction properties.

Corrosion products	Reaction and neutron energy*	Activation cross-section and half-life	γ-ray energy MeV (Intensity %)*
^{24}Na	$^{27}Al(n, a)^{24}Na$ ($E_n > 11.6$ MeV)	6×10^{-8} barn (5.3989×10^4 s)	1.368 (99.994)
	$^{23}Na(n, \gamma)^{24}Na$ (E_n is thermal)	0.53×10^{-4} barn (5.3989×10^4 s)	2.754 (99.855)
^{56}Mn	$^{55}Mn(n, \gamma)^{56}Mn$ (E_n is thermal)	13.4 barn (9.2808×10^3 s)	0.8467 (98.85) 1.8107 (26.9) 2.1131 (14.2) 2.5231 (1.02)
^{59}Fe	$^{58}Fe(n, \gamma)^{59}Fe$ (E_n is thermal)	0.9×10^{-4} barn (1.6018×10^5 s)	0.1426 (1.02) 0.1923 (3.08) 1.0992 (56.5) 1.2916 (43.2)
^{60}Co	$^{59}Co(n, \gamma)^{60}Co$	2×10^{-3} barn (1.6638×10^8 s)	1.1732 (99.85) 1.3325 (99.98)
^{99}Mo	$^{98}Mo(n, \gamma)^{99}Mo$ $E_n > 3.1$ MeV	0.45×10^{-4} barn (2.3751×10^5 s)	0.1811 (6.14) 0.3664 (1.20) 0.7395 (12.26) 0.7779 (4.3)

*Interactive chart of Radionuclides NuDat 2.6, NNDC Databases, Brookhaven National Laboratory (http://www.nndc.bnl.gov/nudat2/chartNuc.jsp).

The time dependent CPA in primary loops of PWRs was carried out for flow rate and power changes using constant corrosion rate models [14]. Program CPAIR was written in FORTRAN-77 and it was developed for assessment of primary coolant activity due to several corrosion products in light water reactors. The program was modified as CPAIR-P for effects of fast as well as slow transients on CPA in coolant. Recently the time dependent corrosion product activity calculations were performed after modifying CPAIR-P code for PWRs with long-term operating cycles using coolant chemistry parameters [11].

In this work we focus on time dependent behavior for CPA in primary circuits of PWRs with extended cycles having flow rate transients and necessary changes were made in CPAIR-P code for nonlinear corrosion rates and pH values. Then for two extended operating cycles the CPA is estimated in the presence of both nonlinear corrosion rates and changes in pH values under flow rate transients.

2. Coolant Chemistry for Extended Cycles

Numerous studies have revealed that operating cycle in light water reactors is extendable to burnup value of 45 MWd/kgU in PWRs. The operating cycles of 18 to 24 month duration show an increase in the capacity factor from 3% to 7% [2] [3] [4]. At the beginning of the cycle for a PWR, 1200 ppm of boron is used in a 12 month cycle. Also initially the pH value of 6.9 is obtained with the help of lithium levels. Initial boron concentration must be higher (~1500 to 1900 ppm) for long operating cycles. This in turns translates into much higher pH values in the coolant. The Enriched Boric Acid reduces the required amount of chemical shim. Therefore, high pH value (7.4) is used in the reactor along with 2 ppm of lithium to keep long operating cycles [4].

The long operating cycles employ this scheme in which the coolant has higher pH in start of the cycle along with high values of boron and lithium concentrations. After that, steadily pH value is elevated to 7.4 and then kept constant till the end of cycle. Various scenarios for pH changes in a typical PWR versus effective full power days are shown as **Figure 1** [1]. The pH variations for long cycles are shown as **Figure 1**. In the first case, Natural Boric Acid (NBA) is used with lithium and pH as 2.2 ppm and 6.9 respectively.

3. Mathematical Model

Using deterministic simulation technique a mathematical model was developed and implemented into a computer code along with a suitable numerical integration method. The time derivative of active concentration is given by [14]:

$$\frac{dn_w}{dt} = \sigma\varphi_\varepsilon N_\omega - \left\{\sum_j \frac{\varepsilon_j Q_j}{V_w} + \sum_k \frac{l_k}{V_w} + \lambda\right\} n_w + \frac{K_p}{V_w} n_p + \frac{K_c}{V_w} n_c \qquad (1)$$

where, σ is the group constant. The effective one-group flux is given by ϕ_ε (#/cm^2·s) and N_w (#/cm^3) represents the concentration of target nuclide in water. The N_w, N_p and N_c have meanings of target concentrations in water, piping and core, in atoms/cm^3. The values of concentrations of the activation

products in primary coolant, piping and core are given as n_w, n_p and n_c respectively, in #/cm³. The sum over *j* for $\varepsilon_j Q_j$ is given by:

$$\sum_j \varepsilon_j Q_j = \varepsilon_I Q_I + \varepsilon_p Q_p + \varepsilon_c Q_c + \varepsilon_F Q_F \qquad (2)$$

where, $\varepsilon_I Q_I$, $\varepsilon_p Q_p$, $\varepsilon_c Q_c$ and $\varepsilon_F Q_F$ represent values of removal rates for ion exchanger, deposition on pipes, core surface depositions and filter based removals, respectively. The primary coolant leakage rate from the *k*th leak is given by l_k and removal rates from piping scale and from core surface are given by K_p and K_c respectively in #/cm²·s. Table 1 gives summary of decay data for various isotope of interest. In this work, experimental values of the removal rates from the published PWR data were used [10]. Some of the values are shown in Table 2 [10] [11].
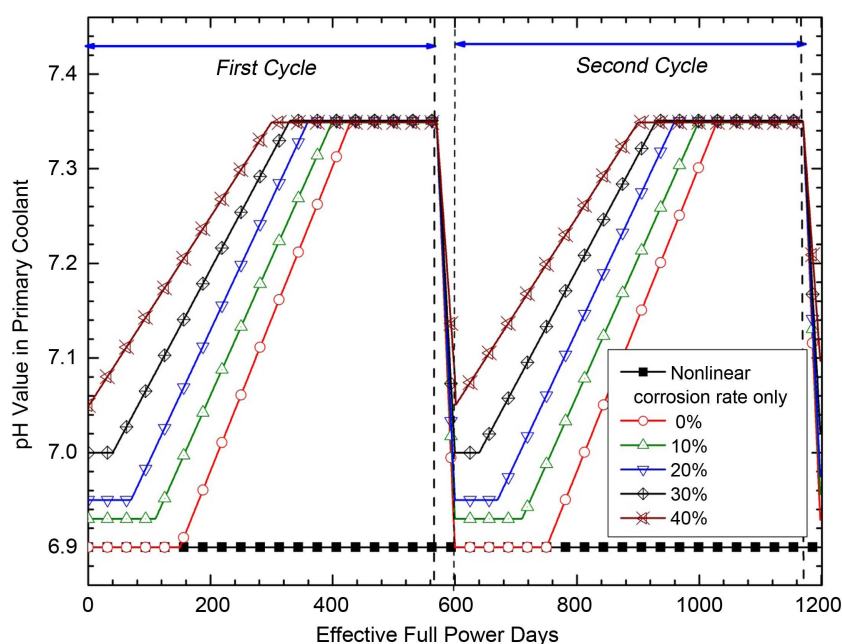

Figure 1. The pH-value in the primary coolant for two cycles as a function of effective full power days (EFPDs) for various indicated values of 10B in boric acid.

Table 2. Experimental values of exchange rates in a typical PWR.

Rate type	Value*
Deposition on core ($\varepsilon_c Q_c$)	80.0 cm³ per second
Deposition on piping ($\varepsilon_p Q_p$)	13.7 cm³ per second
Ion-exchanger removal ($\varepsilon_I Q_I$)	500 to 781 cm³ per second
Re-solution ratio for core (K_c)	40.0 cm³ per second
Re-solution ratio for piping (K_p)	6.9 cm³ per second
Volume of primary coolant (V_w)	1.37×10^7 cm³
Volume of scale on core (V_c)	9.08×10^6 cm³
Volume of scale on piping (V_p)	1.37×10^6 cm³
Total corrosion surface (S)	1.01×10^8 cm²
Average corrosion rate (C_o)	2.4×10^{-13} gm per cm² per s

The temporal variation of dissolved boron in primary coolant are given by a parameter $h(t)$:

$$h(t) = \begin{cases} a_0; \ a_0 \leq 1, \ t_{\min} < t < t_1 \\ 1 + \psi \left\{ ph(\xi, t) - ph(\xi, t_0) \right\}; \ t_1 < t < t_2 \\ b_0; \ b_0 \leq 1, \ t_2 < t < t_{\max} \end{cases} \tag{3}$$

here, ψ represents the slope estimated from the power plant operation data; $ph(\xi, t)$ is boric acid concentration (ξ) and time dependent pH-value; a_0 and b_0 are fractions in the initial and final phases of operation cycle. The residence time of primary coolant in core is represented by T_c while the time taken for circulating once in primary circuit is given by T_L. The temporal variation of concentration of target nuclide can be given as

$$\frac{dN_w}{dt} = -\left\{ \sum_j \frac{\varepsilon_j Q_j}{V_w} + \sum_k \frac{l_k}{V_w} + \sigma \varphi_\varepsilon \right\} N_w + \frac{K_p}{V_w} N_p + \frac{K_c}{V_w} N_c + S_w \tag{4}$$

$$S_w = \frac{C(t) S}{V_w} \frac{N_0}{A} f_n f_s \tag{5}$$

where, N_p and N_c represent the target nuclide concentration on the piping and on the core respectively and S_w represents the corrosion production rate.

The removal rates by ion-exchanger, core deposition and leakage are function of primary coolant flow rate. The corresponding rate of the core scale activity is given by following model:

$$\frac{dn_c}{dt} = \sigma \phi_0 N_c + \frac{\varepsilon_c Q_c}{V_c} n_w - \left\{ \frac{K_c}{V_c} + \lambda \right\} n_c \tag{6}$$

where, the volume of the scale on the reactor core surface is given by V_c (cm³). The ϕ_0 is core averaged thermal neutron flux (*neutrons*/cm²·s). The temporal variation of concentration of target nuclide on the core scale (N_c) is:

$$\frac{dN_c}{dt} = \frac{\varepsilon_c Q_c}{V_c} N_w - \left\{ \frac{K_c}{V_c} + \sigma \phi_0 \right\} N_c \tag{7}$$

The governing equation for the rate of change of active material on the piping scaling (n_p) is given by the following expression:

$$\frac{dn_p}{dt} = \frac{\varepsilon_p Q_p}{V_p} n_w - \left\{ \frac{K_p}{V_p} + \lambda \right\} n_p \tag{8}$$

where, the scale volume on the piping is V_p (cm³). Then, the temporal variation of target nuclide on piping walls (N_p) is

$$\frac{dN_p}{dt} = \frac{\varepsilon_p Q_p}{V_p} N_w - \frac{K_p}{V_p} N_p \tag{9}$$

Using the above system of Equations (1)-(9), the CPAIR-P computer code [14] has been upgraded in the present study to incorporate temporal variations in corrosion rates along with the pH values and chemical shim.

The upgraded version of CPAIR-P code now simulates the CPA incorporating time dependent corrosion rates and primary coolant chemistry. This program

uses the core design parameters to estimate the group constants (Table 3). For this purpose, the LEOPARD [15] code is employed as subroutine in CPAIR-P/PH program. The LEOPARD is a unit cell based computer program using 54 fast & 172 thermal energy groups used for criticality calculations and group constant generations. Subsequently ODMUG program by Thomas and Edlund developed in 1980 was used for computation of detailed spatial group fluxes [16].

4. Results and Discussion

In this work, computer simulations were done for a typical PWR with a clean system having none impurities [17]. The fractional exchange and re-solution rates were taken from the experimental data for the typical PWR. These experimental values of deposition and re-solution rates are given in Table 2. Also, the values of plant specific data are given as Table 3 [10] [11]. The corrosion prone plant surface area of 10^8 cm^2 is in contact with the primary coolant. Also, in recent work [8] it was shown that corrosion rate approaches an equilibrium value of 2.4×10^{-13} g/cm^2-s. Such a PWR has coolant volume equal to 1.3×10^7 cm^3. In this work we have used the same for the simulations. Figure 2 gives algorithmic details of this program in the form of schematic flow chart.

In first part, the equilibrium value of removal rate of CPA by the ion-exchanger ($\varepsilon_I Q_I$) was found taking a constant value of corrosion rate and the details of methodology can be found in the Reference 14 & 17 respectively. These simulations indicate that the final value for CPA becomes constant when $\varepsilon_I Q_I$ becomes higher than 400 cm^3/s. Consequently, the value of 600 cm^3 per second was used for removal rate in this work. Behavior of dominant corrosion

Table 3. Typical design specifications of a PWR.

| 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
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}%)	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
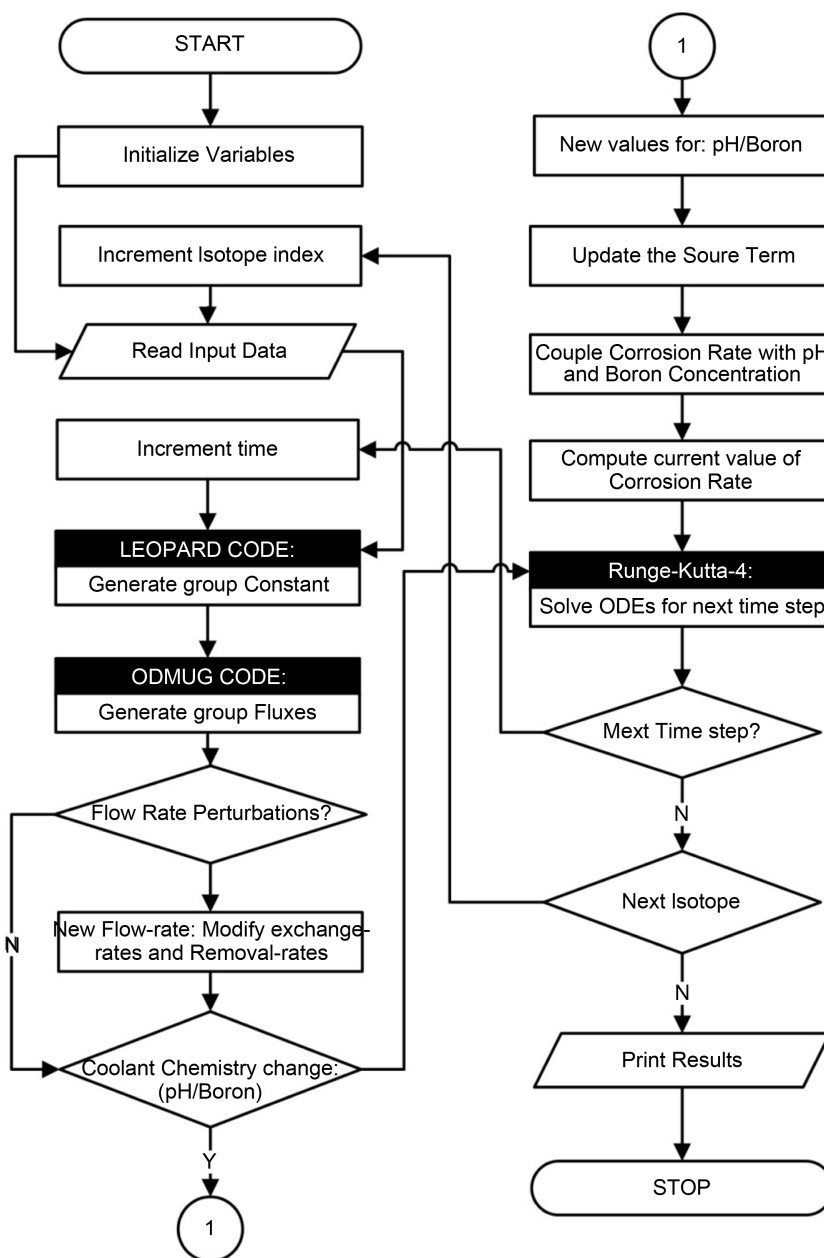
Figure 2. Logical flow chart of the computer modified program CPAIR-P/PH.

isotopes (^{56}Mn, and ^{60}Co) was observed and we saw that when reactor is operating the main contribution comes from ^{56}Mn and after the shutdown the cobalt isotopes dominate the activity.

4.1. Time Dependent pH Value

Typical behavior of the pH-values for two cycles as a function of time is shown as **Figure 1**. Here, different curves indicate different levels of ^{10}B in mixed in the chemical shim. When mixed B-10 is high the pH-value becomes also high in the system. Figure shows that the pH level gradually increases from lower to higher values and it saturates to about 7.35 in both first and second cycle. Increase in pH value lowers the solubility of corrosion products in the coolant. Also high pH

results in a positive temperature coefficient of solubility and this reduces the transport of corrosion products. Also, any increase in the operation length will affect the generation of corrosion product. In this work, we have assumed a constant corrosion rate during the steady state operation.

4.2. Nonlinear Changes in Corrosion and pH Effects

PWRs have in general nonlinear changes in the corrosion rate during their operation. This is in addition to the coolant chemistry effects. The following mathematical model is used for the first cycle:

$$C(t) = \begin{cases} 0, & 0 < t < a \\ h(t)C_s\left\{1 - \exp\left(-m(t-a)\right)\right\}, & a \leq t \leq b \\ h(t)f_{cs}C_s, & b < t \leq c \end{cases} \quad (10)$$

The variable m represents slope having a constant value in the time interval $a \leq t \leq b$. The variable C_s represents the optimum value of rate attained for $t > b$.

Similarly for the second cycle, the initial corrosion rate is non-zero because of a residual value from the first cycle which is enhanced by an increase in corrosion rate along with a opposing decrease due to effects of the pH-value. It is mathematically modeled by the following expression:

$$C(t) = \begin{cases} h(t)\left[f_{cs}C_s + C_s\left\{1 - \exp\left(-m(t-c)\right)\right\}\right], & c \leq t \leq d \\ h(t)f_{cs}\left[C_s + f_{cs}C_s\right], & d \leq t \leq e \end{cases} \quad (11)$$

The values of 600, 1170 and 1200 days were used for *c*, *d*, and *e* respectively. The activity was then determined from the modified program CPAIR-P for both cycles. These simulations were carried out for various boron concentration scenarios. When $h(t) = 1$, the effect of pH-value is nominal and saturation values are largest of all cases. During first cycle, saturation values for Co-60 are 0.24 μCi/cm³ (core region), 0.10 μCi/cm³ (primary pipes) and 0.016 μCi/cm³ (primary coolant).

4.3. Flow Rate Perturbations

When the cross-sectional area for flow changes and/or the primary pump speed gets reduced then the flow rate transient occurs in the reactor. As a result the fuel, moderator and coolant temperatures change and start affecting the neutron flux. This further modifies the production of activated corrosion products and their loss terms. In this work, we have introduced a flow rate perturbation taking PWR operating at full power and the activated products has reached to their saturation values. Here we have not allowed the reactor to scram during these simulations to study the consequences of these simulations. Using the modified CPAIR-P program, the corrosion product activity was estimated in the coolant, primary pipe scale and in the reactor core. Flow rate changes were assumed to be linear and were started at time, t_{in} and remained effective for t_{max}-t_{in} time period. Such a model is introduced by the parameter $g(t)$:

$$g(t) = \begin{cases} 1, & t < t_{in} \\ 1 - \alpha(t_{in} - t), & t_{in} \leq t \leq t_{max} \\ w_f/w_0, & t > t_{max} \end{cases} \tag{12}$$

Flow rate decrease has a slope of α and t_{in} is the time of initiation of mass flow rate perturbation.

The mass flow rate perturbation is initiated at $t_0 = 300$ d for several $\Delta w = w_f - w_0$ values. In time period of ten days the flow rate decreases from w_0 to w_f. The reactor remains at full power and it is not allowed to undergo a scram. Simulation results for first and second cycle are shown as **Figure 3** for activity due to ^{60}Co for 40% ^{10}B in chemical shim. The results for the first cycle indicate that CPA approaches a higher final value. The new saturation value keeps on increasing when $\Delta w = w_f - w_0$ is increased. If the flow rate is allowed to change, Δw is varied from 0 to 0.8 w_0. Then, the new saturation activity in coolant increases from 0.011 µCi/cm³ to 0.017 µCi/cm³. When the ion-exchanger purification rate, $\varepsilon_I Q_I$, becomes low than these saturation value will increase further. Also, an increased deposition, re-solution and leakage can also get increased in these transients. Similarly, for the second cycle the simulations predict higher peak values in comparison with peak values in the first cycle (**Figure 3**). Also, the saturation values in the second operating cycle are about 14% higher as compared with the values in the first cycle. These results indicate that flow rate perturbations substantially affect the balance of CPA in PWRs and their effects propagate in next operating cycles.

5. Conclusion

The CPA under flow rate perturbations was investigated in primary loop of a
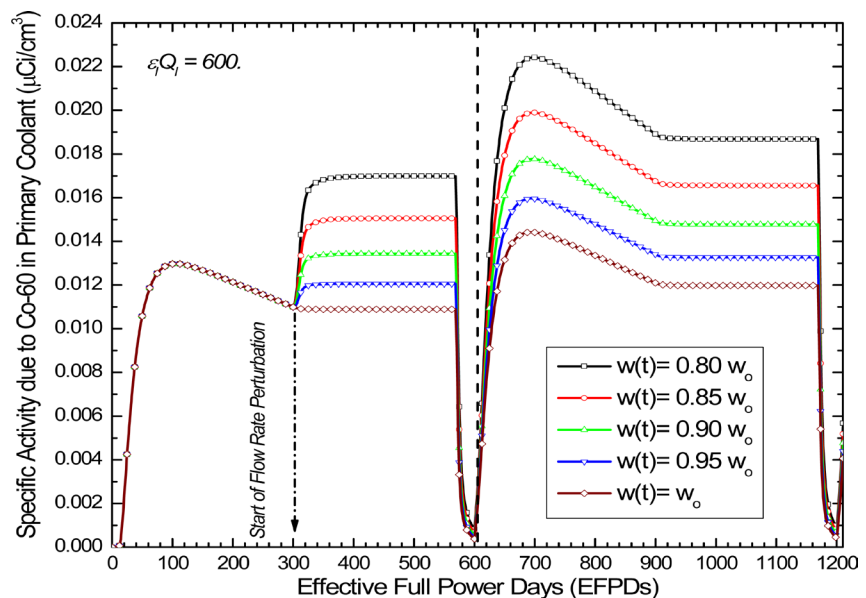

Figure 3. Comparison of specific activity due to 60 Co in the primary coolant under flow rate perturbation having nonlinearly rising corrosion rate and 40% 10B in dissolved boric acid. Flow rate perturbation is introduced at 300 day for $\Delta t = 10$ days.

typical PWR with long operating cycle. These cycles affect the primary coolant chemistry in terms of operational levels of boron and optimum pH values. These simulations indicate that for the normal reactor operation conditions the values of specific CPA approach equilibrium value fairly quickly. The pH values, removal rates and cycle length affect the saturation level of CPA in the system. The CPA levels have been compared for flow rate transients in the presence of higher levels of ^{10}B in chemical shim. When the flow rate transient is applied, the saturation values of CPA go up. If the flow rate is allowed to change, Δw is varied from 0 to 0.8 w_0. Then, the new saturation level in the coolant increases from 0.011 μCi/cm³ to 0.017 μCi/cm³. The saturation values in the second operating cycle are about 12% higher when compared with values in the first cycle.

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