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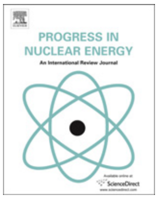

Time-dependent corrosion product activity in a typical PWR due to changes in coolant chemistry for long-term fuel cycles

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ABSTRACT

Extended fuel cycles with 18–24-month duration have complicated effects on primary coolant chemistry and finding optimum pH concentration range is difficult. For extended burnup cycles, study of changes in coolant activity due to corrosion products have been carried out for a typical pressurized water reactor (PWR) under pH and boric acid variations for linearly as well as nonlinearly accelerating corrosion rates. The computer program CPAIR-P (Deeba et al., 1999) has been modified to accommodate for time-dependent corrosion coupled with the effects of coolant chemistry. Results for ^{24}Na, ^{56}Mn, ^{59}Fe, ^{60}Co and ^{99}Mo show that the specific activity in primary loop approaches equilibrium value under normal operating conditions fairly rapidly. During reactor operation, predominant corrosion product activity is due to ^{56}Mn and after shutdown cobalt activity dominates. These simulations suggest that the effect of increase in pH value for an extended 24-month cycle on specific activity in the form of a decrease in the activity smeared by a linearly rising corrosion. The new saturation values for activity at the end of cycle are lower ($\sim$50%) than a reactor operated at constant low pH/natural boric acid in coolant. For linearly accelerated corrosion and an increase in pH value, the coolant activity shows an initial rise and then it falls to a lower saturation level at the end of cycle when pH becomes large and the activity follows the slope of corrosion rate. For nonlinear rise in corrosion rate coupled with the pH rise from 6.9 to 7.4 and use of enriched boric acid (20%–40%) results in coolant activity peak during the cycle and it also approaches much smaller saturation values at the end of cycle when compared with the activity for the system having constant low pH value (6.9) in the coolant. In this paper we show that the use of enriched boric acid as chemical shim actually lowers the primary coolant activity when higher pH values are employed in coolant rather than natural boric acid. For multiple long-term fuel cycles the corrosion product activity shows an initial rise to a maximum value and then it fall back to low saturation values due to high pH concentration for enriched boric acid (40%) as chemical shim. If the removal rates of ion-exchanger are low ($\varepsilon_I Q_I \sim 100$–300), then the activity builds up. However, when ion-exchanger performs well ($\varepsilon_I Q_I \sim 600$–900) then the overall cobalt activity remains low and saturation value does not grow much with cycles.

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1. Introduction

For past twenty years the discharge burnups of spent fuel from Pressurized Water Reactors (PWRs) have been substantially increased to 40–50 MWd/kgU as compared to the previous generations having average burnup of 33 MWd/kgU. Such long-term fuel cycles need higher initial enrichments and/or higher fuel loading and better neutron absorption at the beginning of cycles. Long-term safety means increase in the soluble boron concentration in the primary circuit and changes in lithium concentrations to optimize the reactor kinetic conditions. Moreover, it was also perceived that corrosion products were mostly composed of magnetite (Fe_3O_4) in the primary loop. However, recent studies have shown that the corrosion products mainly are nickel-ferrite ($Ni_xFe_{3-x}O_4$) (Robertson, 1989). Therefore for such systems, higher pH levels of 7.4 are more suited as compared to pH value of 6.9 for primary coolant. It is also observed that corrosion product activity can be minimized using high pH values. Moreover, a typical PWR uses natural boric acid (NBA) as soluble poison or chemical shim. Several studies have shown that enriched boric acid (EBA) reduces the needed concentration and an elevated pH of 7.4 can be achieved by using an acceptable 2.2 ppm of lithium without corrosion cracking problems (EPRI, 1992; KAERI, 1995).

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Various studies have shown that water becomes very corrosive at high values temperatures and pressures present in PWRs. The decomposition of water by radiation increases its corrosive nature. The corrosion products may originate as soluble and insoluble oxides, or in other particulate forms. The rate of corrosion in the reactor primary system keeps on increasing as the time of reactor operation at full power increases (Hirschberg et al., 1999; Varga et al., 2001).

In the reactor core, corrosion products become activated due to neutron irradiation and their subsequent buildup in the core, in the primary coolant and on piping inner surfaces takes place due to their long life and exposure. Their sizeable radioactivity can prohibit access to primary pumps, valves and its vicinity. The buildup of activity in the circulating coolant, on the coolant piping inner surfaces and on the core surfaces does pose problems to the reactor operation and maintenance in terms of accessibility demands (Mirza et al., 1998, 2003; Deeba et al., 1999; Rafique et al., 2005).

In PWRs the corrosion product activity is primarily due to short-lived ^{56}Mn and ^{24}Na. Nearly all the long-lived activity in the coolant is due to iron, molybdenum and cobalt with most significant radionuclides as ^{59}Fe, ^{99}Mo and ^{60}Co. Various nuclear properties of these nuclides are summarized as Table 1 (Deeba et al., 1999). The ^{55}Mn has an activation cross-section of 13.4 b for the thermal neutrons to produce ^{56}Mn. The neutron activation of structural ^{27}Al and activation of ^{23}Na from salt impurities in water can produce ^{24}Na. The use of high-purity water, demineralization of water and the presence of filters keep the amount of dissolved salts to less than 0.05 ppm (Mirza et al., 2003). However, the half-lives of the dominant corrosion products are longer than two hours. Therefore, the primary coolant retains the activity for several hours even after the reactor shutdown and any transient condition during operation can further increase the end of cycle coolant activity.

For the reactor coolant corrosion product experimental data there are problems in withdrawing representative samples from PWR primary coolant through long sampling lines. It has been suggested in various studies that measured concentrations of some corrosion products (e.g., soluble Co-60, Co-58 and Mn-54) are strongly dependent on sampling flow rate and pH values and born concentration (Bergmann and Roesmer, 1984; Kang and Sejvar, 1985; EPRI, 1992).

Operating parameters of the reactor also strongly affect the types of radionuclides formed, the levels of saturation activity reached and the rates at which the saturation is reached. These include the composition of the materials in contact with the coolant, the amount and the types of the impurities present in the coolant, reactor power, residence time of coolant in core, temperatures, pressure, coolant flow rates, corrosion rates, filter efficiency and deposition rates of radioactive elements in coolant.

Table 1
Activation products and their reaction properties.[a]

Corrosion products	Reaction and neutron energy	Activation cross-section and half-life	γ-ray energy
^{24}Na	^{27}Al$(n, \alpha)^{24}$Na ($E_n > 11.6$ MeV) ^{23}Na$(n, \gamma)^{24}$Na (E_n is thermal)	6×10^{-4} b (15.4 h) 0.53 b (15.4 h)	4.1 MeV
^{56}Mn	^{55}Mn$(n, \gamma)^{56}$Mn (E_n is thermal)	13.4 b (2.58 h)	2.13 MeV (15%) 1.81 MeV (24%)
^{59}Fe	^{58}Fe$(n, \gamma)^{59}$Fe (E_n is thermal)	0.9 b 45.1 h	1.17 MeV (99.9%) 1.33 MeV (99.9%)
^{60}Co	^{59}Co$(n, \gamma)^{60}$Co	20 b (5.3 years)	1.173 MeV (99.9%) 1.332 MeV (99.9%)
^{99}Mo	^{98}Mo$(n, \gamma)^{99}$Mo ($E_n > 3.1$ MeV)	0.45 b (67.0 h)	0.78 MeV (8%) 0.74 MeV (8%)

[a] Deeba et al., 1999.

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

Rate type	Value
Deposition on core ($\varepsilon_c Q_c$)	80.0 cm^3 per s
Deposition on piping ($\varepsilon_p Q_p$)	13.7 cm^3 per s
Ion-exchanger removal ($\varepsilon_I Q_I$)	500–781 cm^3 per s
Re-solution ratio for core (K_c)	40.0 cm^3 per s
Re-solution ratio for piping (K_p)	6.9 cm^3 per s
Volume of primary coolant (V_w)	1.37×10^7 cm^3
Volume of scale on core (V_c)	9.08×10^6 cm^3
Volume of scale on piping (V_p):	1.37×10^6 cm^3
Total corrosion surface (S)	1.01×10^8 cm^2
Average corrosion rate (C_o)	2.4×10^{-13} g per cm^2 per s

[a] Rafique et al., 2005 and Mirza et al., 2005.

During past fifteen years, series of studies were done on coolant activation in reactors and effects of flow rate and power perturbations remained in focus. It was seen in simulations of low and high flux systems that transients under reactivity and loss of flow lead to peaking of neutron flux in reactor and production of activity in coolant (Iqbal et al., 1997; Mirza et al., 1998). These strongly affect the coolant activity and corrosion rate. Void coefficient, Doppler coefficient and temperature of moderator showed significant effects on power peaking in a non-uniform manner within the core.

Venz and Weidmann showed that for extended fuel cycles in PWRs, the reactor coolant chemistry plays an important role and dose at nuclear power plant Beznau reduces in the primary coolant circuit when pH level was increased to 7.4 (Venz and Weidmann, 1996). Also, the evaluation of corrosion products activity in PWRs as a function of variations in the primary coolant chemistry for long-term fuel cycle was done by Song and Lee (2003). They used COTRAN code based on a two-layer model and predicted that the generation of the CRUD increases as the operation cycle duration is increased. They also predicted that enriched boric acids in coolant reduce the amount of CRUD and high pH operation periods become possible. The results were preliminary and they suggested extensive studies to analyze the real reactor CURD behaviors.

Modeling of time-dependent radioactivity due to corrosion products in primary coolants of PWRs under various perturbations were done to investigate effects due to flow coast down and linear decrement of flow (Deeba et al., 1999) for constant corrosion rates. A computer code CPAIR was developed in FORTRAN-77 to calculate specific activity due to ^{24}Na, ^{56}Mn, ^{59}Fe, ^{60}Co and ^{99}Mo in primary circuits of light water reactors. It was shown that minimum value of coolant activity depends strongly on the slope of linear decrement of flow rate. The program was further improved to incorporate the effect of power perturbations on corrosion product activity in coolants of a PWR (Deeba et al., 1999). The computer code was modified as CPAIR-P and effects of fast and slow transients were studied on dose rates due to corrosion activity in coolant. All these studies assumed a constant and uniform corrosion rate and fixed pH values during and after transients. However, the corrosion rate does increase slowly with plant operation; it also increases with temperature and pressure. The rate of increase of corrosion depends on integrated effect of neutron flux, reactor operation cycle length, reactor temperatures and coolant chemistry.

This work aims at simulating the behavior of corrosion product activity in a typical PWR for extended cycles when coupled effects of both an increase in the corrosion rate due to normal processes happen and a decrease due to coolant chemistry is present in the primary circuit respectively. These changes are superimposed and the computer program CPAIR (Deeba et al., 1999) is modified to incorporate both the linear and nonlinear rise in the corrosion rates and effects of pH/boron concentrations in the coolant. First, we

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