Journal of the Korean Nuclear Society
Volume 24, Number 1, March 1992

Evaluation of Primary Coolant pH Operation Methods for the Domestic PWRs

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(Received July 1, 1991)

국내 PWR의 일차냉각재 pH 운전방법의 평가

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Abstract

Radioactive nuclides deposited on out-of-core surface after the radiation in the core by the transport of corrosion products (CRUD) through the primary coolant system in PWR which is the major plant type in Korea, are leading sources of radiation exposure to plant maintenance personnel. Thus, the optimal chemistry operation method is required for the reduction of radiation exposure by the corrosion products.

This study analysed the actual water chemistry operation data of four operating domestic PWRs. And in order to evaluate the coolant chemistry operation data, a computer code which can calculate the activity buildup in the various chemistry conditions of PWR coolant was employed. Through the analysis of comparison between the activity buildup of actual water chemistry operation mode and that of assumed Elevated Li operation mode calculated by the computer code, it was found that the out-of-core radioactivity can be reduced by diminishing the deposition of corrosion products on the core in case that the Elevated Li operation mode is applied to the coolant chemistry operation of PWR. And the higher coolant pH operation was shown to have the advantage of the reduction of out-of-core activity buildup if the integrity of system structural materials and fuel cladding is guaranteed.

요 약

국내 원자력 발전소의 주요 기종인 가압 경수로에서는 일차 냉각계통을 통한 부식생성물 (CRUD)의 이동에 의해 노심에서 방사화된 후 노외표면에 침적된 방사성 핵종은 원전 종사자 방사선 피폭의 주원인이 된다. 따라서, 부식생성물에 의한 방사선 피폭을 감소시키기 위한 최적 화학운전 방안이 요망된다.

본연구에서는 운전중인 국내 4개 발전소의 실제 수화학 운전 자료를 분석하였으며, 냉각재 화학 운전 자료를 평가하기 위해 냉각재 수화학 조건에 따라 방사능 생성양을 계산할 수 있는 Computer 코드를 이용하였다. 실제 수화학 운전조건과 가정된 Elevated Li 운전조건에 따른 운전결과를 Computer 코드에 의해 예측하여 비교한 결과, Elevated Li 수화학 운전방법을 적용할 경우, 현재 적용되는 수화학 운전방법에 비하여 노심에서 부식생성물의 침적을 감소시킴으로써 노외 방사능 양을 상당히 감소시킬 수 있음을 알았다. 또한 계통 구성재질과 핵연료봉의 건전성이 보장되는 한 냉각재 pH를 상승시키면 노외 방사능 생성감소에 유리함을 밝혔다.

1. Introduction

The minimization of radiation levels is a frequently discussed issue in the Nuclear Industry. Radioactive nuclides deposited on the out-of-core surface in PWR primary coolant system, caused by the transport of corrosion products(crud) through the primary coolant, are a major source of radiation exposures to plant maintenance personnel. The main contributors to dose rate buildup in the primary side of PWRs are Co^{60}, which is an activation product of Co^{59} formed by neutron capture, and Co^{58}, which is formed from the Ni^{58} by an (n, p)-reaction. Both nickel and cobalt are released from the out-of-core surface by corrosion effect and transported to the core, where the activation can take place. They are also released directly from in-core surface as activation products, again by corrosion effect/1/. Judging from these processes of corrosion products in the primary coolant system, optimization of the PWR primary side water chemistry is recommended as the attractive choice of minimizing the out-of-core radiation buildup.

Modeling and numerical estimations of crud and activity transport are essential to systematic approaches to the optimization of the PWR primary water coolant chemistry. Many investigators have developed the computer codes such as CORA, CRUDSIM, and PACTOLE/2,3,4/, in order to estimate the effect of coolant chemistry(pH control) on out-of-core activity builup.

But, little research on the radiation field buildup related to water chemistry operation method has been done in Korea. And the domestic plants have been operated according to the operation method only given by the vendors up to now. Therefore, this study focuses on the evaluation of the actual primary water chemistry operation data for the domestic PWRs using the concept of CRUDSIM model because of its various attractive features of data analysis and conceptual simplicity of application. And it is our belief that this work will more or less contribute to the establishment of the optimum water chemistry operation method for the primary coolant of domestic PWRs by predicting activity buildup according to the various operation methods and judging the operation methods through the comparison of the predicted data.

2. Role of coolant chemistry

PWR plant radiation fields can be affected by numerous factors including type of plant operation, shutdown procedure, operational chemistry control, plant configuration and construction, and time of operation/5,6/. Although many operating and design factors probably impact significantly on radiation fields in PWRs, in fact, the only parameter which the plant chemist can vary in a controlled manner is the coolant lithium concentration, that is, the coolant pH/7/.

The basis for coolant pH control is the requirement to minimize transport of corrosion products around the circuit. Until recently, the pH for the primary coolant at 300℃ has been recommended

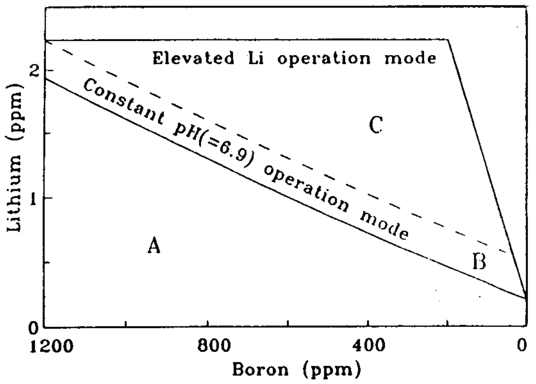

Fig. 1. Boron–Lithium Operating Regions.

as 6.9 (area B in Fig.1). This corresponds to the pH with zero temperature coefficient of solubility of magnetite which was considered as major components of corrosion products in the primary coolant of PWR/8/. The solubility data for magnetite were initially used to choose the optimum pH and to model the transport of corrosion products. But, many researchers have lately reported that deposits on fuel assemblies have a nickel ferrite structure of varying stoichiometry/9/. And they showed the solubility of nickel ferrite along its several compositions as a function of pH, temperature and dissolved hydrogen concentration/10/. These results indicate that a pH higher than 6.9 is required for favorable (zero or positive) temperature coefficients of solubility of nickel ferrite in the coolant at 300℃. Even though EPRI recommended the Elevated Li operation mode (area C), there is yet no consensus concerning for the optimum pH operation method around the world. However, it has been suggested in many papers that the lithium and boron chemistry should be coordinated to achieve a constant or increasing pH in the range 6.9 to 7.4 (area B,C) and that operation at a pH below 6.9 (area A) is not recommended.

Several decades have been passed since the first unclear power plant started operation. But, unfortunately the optimum pH range has not yet been established/11/. Therefore, before the application of recently recommended various operation modes to the domestic PWRs, it is very important to investigate the pH operation histories of domestic PWRs and to evaluate the radiation field buildup according to the variations of pH operation mode.

3. Modeling of CRUD Transport

From the utility stand point, an activity transport computer code is desirable for two main quantification tasks ; a) the forward prediction of plant dose development and b) the prediction of the effect on plant dose of operational and design modifications. A comprehensive review of the codes is available elsewhere /2,3,4/. CORA, CRUDSIM and PACTOLE computer codes have been well known for prediction of radiation field buildup caused by activity transport in the primary coolant system of PWR.

The CORA computer code/2/ developed by Westinghouse. The various transport mechanisms of the CORA code are modeled semi–empirically between the individual nodes, which represent homogeneous sources and sinks of the corrosion products in a PWR primary system. The PACTOLE code/4/, developed by CEA in France, considers 20 different zones in the PWR coolant circuit. The governing parameter is the solubility of system materials.

It should be noted that the comprehensive modeling in CORA and PACTOLE may obscure the real contribution of dominant mechanism, if any. Even though each computer code is continuously updated to match new plant data or experimental results, it is not clear that the true significance of each mechanism has been appropriately accounted for/12/.

The semi–empirical CRUDSIM code/3/ employs a conceptually simple model compared with the comprehensive models of CORA and PACTOLE. In this model, only two nodes represent the PWR primary coolant circuit and the radioactive elements considered are Co^{58} and Co^{60}. Crud and activity transport are predicted only on the basis of a driving force of solubility difference. Therefore, the results of CRUDSIM calculations are generally used in relative or empirically adjusted comparisons of various coolant chemistry conditions (i.e., pH). Reasonable comparisons with measured plant histories can be obtained using a judiciously selected transport characterization parameters. Because of the attractive features of good results and conceptual simplicity, we selected the CRUDSIM ("Slurry tank") model as the basic frame of our computer code for the evaluation of the domestic PWRs water chemistry operation data. However, we employed the semi-empirical equation for solubility calculation recently published by Lee and Lindsay/12,15/ since the solubility is very important for the driving force of crud transport in the model. And we also utilized the various plant specific data such as coolant temperature, heat transfer area of core and steam generator and coolant flow rate for each plant to apply our computer code to the domestic plants.

The model is illustrated in Figure2. In this model, the hot tank represents the in-core part of the primary system where the radiation occurs, while the cold tank represents the entire out-of-core part of the system including steam generator. Corrosion products enter the cold tank at a constant rate R, measured in kg Fe/day. Coolant is taken from the bottom of cold tank at primary coolant flow rate, F(kgH_2O/day), and returned to the top of the tank at the same rate. A fraction of this flow, β F passes through hot tank and is equilibrated with the solid contents of hot tank. The remainder, $(1-\beta)$ F, is returned directly to the top of cold tank.

The selection of coolant temperature influences on the overall results greatly. The cold tank and hot tank temperature (t_c and t_h) in the CRUDSIM code were 285℃ and 320℃ respectively. However, in this work we selected the t_c and t_h from FSARs of each plant and correlated t_h with t_c as equation (1):

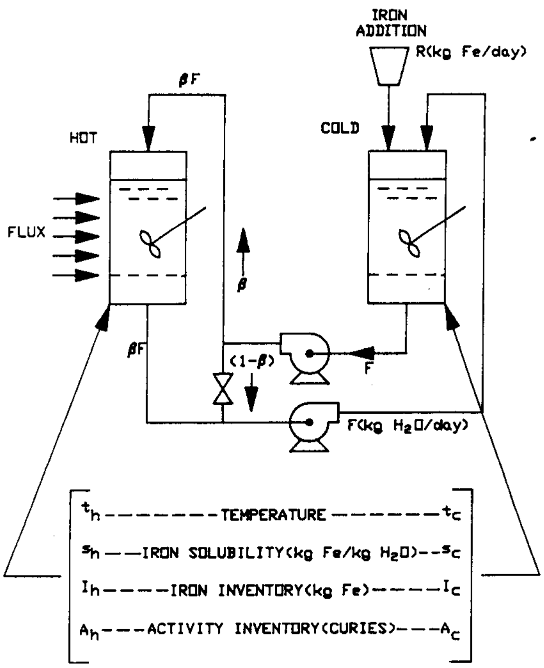

Fig. 2. Schematic Representation of CRUDSIM("Slurry Tank") Model.

$$t_h = t_c + a' P \tag{1}$$

Here, a' is the plant specific values and P is percent of full power. Thus, the temperature selection in our code describes the real temperatures of each plant more closely than in the CRUDSIM code.

In accord with the assumptions of the model, the material balances in hot and cold tanks can be described as follows:

$$\frac{\Delta I_h}{\Delta \theta} = \beta F S_c - \beta F S_h \tag{2}$$

$$\frac{\Delta I_c}{\Delta \theta} = [\ (1-\beta)F\ S_c + \beta F\ S_h\ ] - F\ S_c + R \tag{3}$$

Here, S_c is solubility of iron (kg Fe/kg H_2O) in the cold tank, S_h is iron solubility in the hot tank.

With isotopic exchange in each tank, the activity balances in hot and cold tanks for Co^{60} can be obtained as follows:

$$\frac{\Delta A_h^{60}}{\Delta \theta} = \alpha^{60} P I_h + \beta F S_c \frac{A_c^{60}}{I_c} - \beta F S_h \frac{A_h^{60}}{I_h} - \lambda^{60} A_h^{60} \tag{4}$$

$$\frac{\Delta A_c^{60}}{\Delta \theta} = \beta F S_h \frac{A_h^{60}}{I_h} - \beta F S_c \frac{A_c^{60}}{I_c} - \lambda^{60} A_c^{60} \tag{5}$$

Here, λ^{60} is the decay constant for Co^{60} and α^{60} is a production rate constant for Co^{60}. A similar set of equations is derived for Co^{58}.

We calculated the iron solubilities from pH, temperature and dissolved hydrogen concentration which are given according to the operating conditions of the primary coolant. This solubility calculation is considered as the most important step since the crud and activity transport is calculated only on the basis of a driving force of solubility difference in this computer code. CRUDSIM code used the data of solubility experimental results by Sandler and Kunig/13,14/. But in this study, we utilized the equation and parameters recently published by Lee and Lindsay/12,15/. The model equation is,

$$S = 10^6 A P_{H2}^{1/(3-x)} \left(K_0 \frac{z^2}{\gamma_1^2} + K_1 z + \frac{K_2}{\gamma_0} + \frac{K_3}{\gamma_1^2 z} \right) \tag{6}$$

where,

S = iron solubility

A = stoichiometric factor for the solid = $(1-x)^{(1-x)/(3-x)}$

z = hydrogen ion concentration, given by $z = 10^{-pH}$

P_{H2} = hydrogen partial pressure (atm)

γ_1 = generic activity coefficient for univalent ions

γ_0 = activity coefficient for neutron species

and the coefficient $K_0 \cdots K_3$ are given by

$\log_{10}K_0 = A_0 + B_0/T$

$\log_{10}K_1 = A_1 + B_1/T$

$\log_{10}K_2 = A_2 + B_2/T$

$\log_{10}K_3 = A_3 + B_3/T + \log_{10}K_w$

with K_w = thermodynamic ion product for water

T = absolut temperature(°K)

The parameters $A_0 \cdots A_3$ and $B_0 \cdots B_3$ are thermodynamic quantities to be determined by least squares analysis. These parameters were obtained through the analysis of experimental data by Westinghouse and CEA based on the iron solubility of non-stoichiometric nickel ferrite published. These calculated values may be considered to be the precisest ones of the iron solubility up to now/12,15/. Details of this program are described elsewhere/16/.

4. Results and Discussions

4.1. The Histories of Coolant Chemistry for Domestic PWRs

Domestic nuclear power plant operation has over 10 years of history but the operation has been done without consideration of relationships between water chemistry and radiation field buildup up to now. In 1986, Westinghouse suggested the Coordinated B–Li operation mode (constant pH mode) and domestic PWRs operation has followed this mode since then.

In order to evaluate the effect of domestic PWR primary coolant chemistry conditions on the plant personnel exposures, water chemistry data were collected from operating PWRs (Kori Unit 1 and 4,

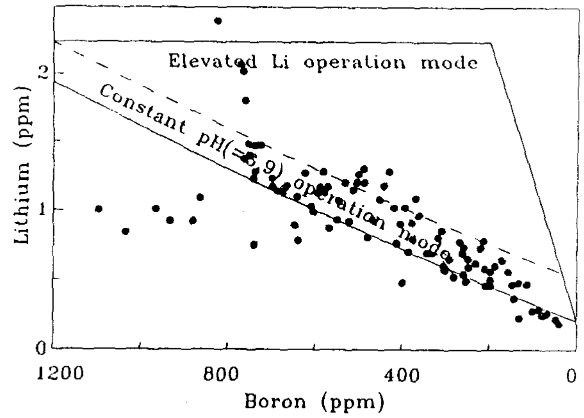

Fig. 3. Boron vs. Lithium Concentrations in KORI Unit 4 Reactor Coolant during Cycle 1.

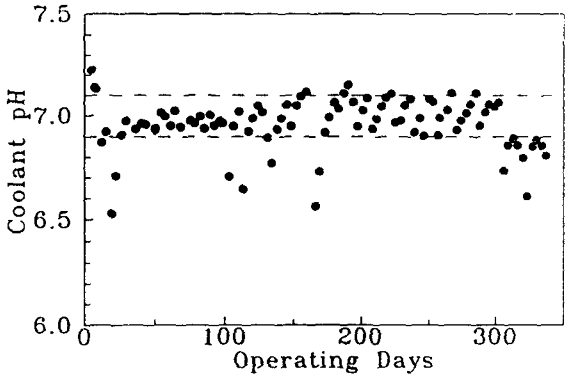

Fig. 4. pH Values of Operation Cycle 1 at KORI Unit 4

Young Gwang(YGN) Unit 1, and Uljin Unit 1) and compared with predicted values by our computer code under the various operation modes.

In Frgure 3, actual B–Li operation data at KORI Unit 4 during cycle 1 were illustrated and compared with predicted values under the coordinated B–Li mode based on the solubility of magnetite and under the Elevated Li mode based on non–stoichiometric nickel–ferrite/11/. The former mode has been utilized in domestic power plants and the latter mode has recently recommended by EPRI. From the Figure 3, we can evaluate whether the plant has been operated according to the suggested pH opeartion mode or not. It is more clearly shown in Figure 4 that the pH of the primary system water at an average coolant temperature had been maintained mostly between 6.9 and 7.1 during 340 days operation of cycle 1 at KORI Unit 4. This result indicates that KORI Unit 4 had been operated very well in accordance with the Coordinated B–Li mode suggested by Westinghouse.

The similiar tendency has been shown in the case of the other plants except KORI Unit 1. Figure 5 shows that the KORI Unit 1 had been operated in the lower pH and irregular mode during cycle 1. This discrepancy of operation mode from the recommended one might be due to the factors that the plant was the first commercial nuclear power plant in Korea and that the operators had no experiences at the beginning of its operation.

4. 2. Radiation Field Buildup of Domestic PWRs

Table 1 describes the calculational results of radiation field buildup in domestic PWRs. Since operation of the KORI Unit 1 which was the first commercial Nuclear Power Plant in Korea, quite lots of activities are estimated to have produced in early operation. And, with the continuity of plant operation, the activity ratio of Co^{58}/Co^{60} is shown to be reduced, and the activity portion of Co^{60}, which has long half–life, is founded to be increased.

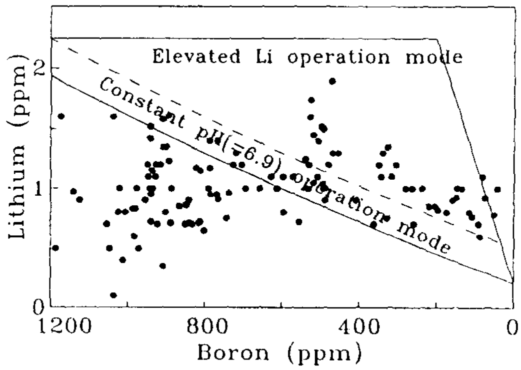

Fig. 5. Boron vs. Lithium Concentrations in KORI Unit 1 Reactor Coolant during Cycle 1.

Table 1. Results of Activity Calculations in Domestic PWRs.

		Ex–Core				In–Core		
Plant	Cycle	Co^{58}	Co^{60}	Iron	$\frac{Co^{58}}{Co^{60}}$	Co^{58}	Co^{60}	Iron
KORI Unit 1	1	5.326	0.802	0.197	6.6	27.20	1.573	0.086
	2	5.472	1.380	0.258	4.0	27.69	1.971	0.087
	3	7.060	1.894	0.302	3.7	35.56	2.366	0.095
	4	7.217	2.201	0.310	3.3	35.23	2.440	0.095
	5	6.960	2.559	0.338	2.7	42.28	3.078	0.107
	6	7.396	2.812	0.343	2.6	44.52	3.273	0.109
	7	7.311	2.983	0.343	2.5	45.83	3.497	0.119
KORI Unit 4	1	3.130	0.297	0.074	10.4	11.61	0.600	0.045
	2	5.214	0.800	0.124	6.5	15.77	1.029	0.058
	3	5.403	1.172	0.153	4.6	21.62	1.445	0.063
YGN Unit 1	1	2.860	0.239	0.073	11.9	7.689	0.363	0.033
	2	4.382	0.607	0.120	7.2	13.14	0.746	0.046
ULJIN Unit 1	1	3.096	0.248	0.072	12.4	7.866	0.378	0.034

* Activity in μCi/cm^2 and Iron in mg/cm^2.

Since our computer code can predict the out-of-core activities according to various chemistry operation methods, it was applied in this work to the evaluation of the chemistry operation history in nuclear power plants and to the establishment of the chemistry operation strategy in future. But it is suggested that these calculation results should not be used in relative comparison between plants because of the difference in operating days, operation conditions (boron, lithium, and dissolved hydrogen concentration), and the days of plant shutdown.

4. 3. Comparison with the Elevated Li Operation Mode

Elevated Li operation mode which was recommended by EPRI recently, maintains the maximum limited lithium concentration (2.2 ppm Li) until reaching a pH of 7.4 (at 300℃) and then follows the pH 7.4 line. In order to evaluate the chemistry factor due to pH related solubility differences only, our calculations were made for the four domestic plants assuming that they had been operated using the Elevated Li chemistry during the first cycle. This was done by using the actual plant concentration of boron and hydrogen, and power values for the first cycle of operation, and by changing only the lithium levels to match the corresponding boron values as if the plant had operated with the Elevated Li chemistry. In Figure 6, the predicted values under the operating condition of the assumed Elevated Li chemistry are compared with actual chemistry operation data in KORI Unit 4 during cycle 1. The relative results are shown in Table 2.

Since the above technique fixed the operation parameters of the coolant cheimstry such as hydrogen concentration and power values, the only effect is changing lithium concentration according to boron concentration of coolant in the plants. It is shown in Table 2 that in the case of Elevated Li mode the amount of activity in the out-of-core can be reduced by 10 to 25% compared with that

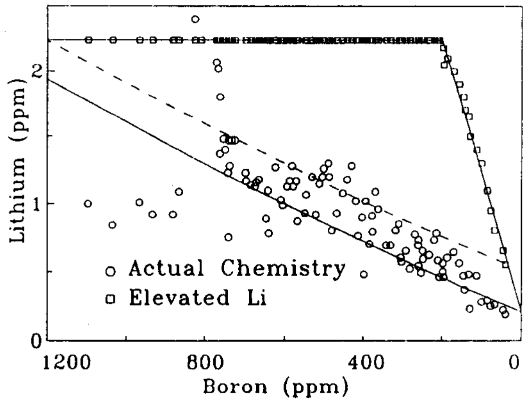

Fig. 6. Assumed Elevated Li Operation Mode Compared with Actual Chemistry Mode in KORI Unit 4 during Cycle 1.

of actual chemistry. But the amounts of iron in the out–of–core are slightly increased. In the case of in–core, both activity and iron are greatly reduced. These results indicate that the Elevated Li mode suppresses the deposition of corrosion products in the core and makes them deposited on the out–of–core. Based on the above results, it is evident that the activation of corrosion products in the core and the amount of the out–of–core radiation buildup can be reduced significantly when the Elevated Li mode is applied.

In Table 3, the data of activity calculation are listed in two cases of operations of actual chemistry mode and assumed Elevated Li mode during cycles 1, 2 and 3 at KORI Unit 4. After cycle 3, the activities in the Elevated Li mode can be reduced by nearly 35% in the out–of–core and by about 75% in–core compared with the ones of actual chemistry mode. These results indicate that we could accomplish more activity reduction if the plant had been operated by Elevated Li mode continuously over entire cycles.

4. 4. Comparison with the Various Operation Modes

We employed the computer code for the evaluation of five lithium–boron operation modes to see the chemistry change effects on the activity buildup. In Figure 7, the concentration change of boron and lithium is shown for the five operation

Table 2. Calculated Relative Activity Values for Domestic PWRs Using Actual Chemistry Data and Assuming Elevated Li Chemistry Operation

Plant	Ex–Core				
	Actual Chemistry		Elevated Li Chemistry		Activity ratio of Elevated Li to Actual
	Activity	Iron	Activity	Iron	
KORI Unit 1	6.128	0.197	4.994	0.207	0.82
KORI Unit 4	3.427	0.074	2.589	0.082	0.76
YGN Unit 1	3.099	0.073	2.792	0.077	0.90
ULJIN Unit 1	3.344	0.072	2.681	0.077	0.80
	In–Core				
KORI Unit 1	28.773	0.856	12.507	0.050	0.44
KORI Unit 4	12.210	0.045	2.701	0.018	0.22
YGN Unit 1	8.052	0.033	2.798	0.017	0.34
ULJIN Unit 1	8.244	0.034	2.791	0.018	0.34

* Activity in $\mu Ci/cm^2$ and Iron in mg/cm^2.

Table 3. Calculated Relative Activity Values for KORI Unit 4 during Cycle 1, 2 and 3 Using Actual Chemistry Data and Assuming Elevated Li Chemistry Operation.

KORI Unit 4	Ex-Core				
	Actual Chemistry		Elevated Li Chemistry		Activity ratio of Elevated Li to Actual
	Activity	Iron	Activity	Iron	
Cycle 1	3.427	0.074	2.589	0.082	0.76
2	6.014	0.124	3.745	0.134	0.62
3	6.575	0.153	4.344	0.163	0.66
	In-Core				
Cycle 1	12.210	0.045	2.701	0.018	0.22
2	16.799	0.058	3.777	0.025	0.23
3	23.065	0.063	6.009	0.029	0.26

* Activity in $\mu Ci/cm^2$ and Iron in mg/cm^2.

* Used actual chemistry values for KORI Unit 4 cycle 1,2 and 3.

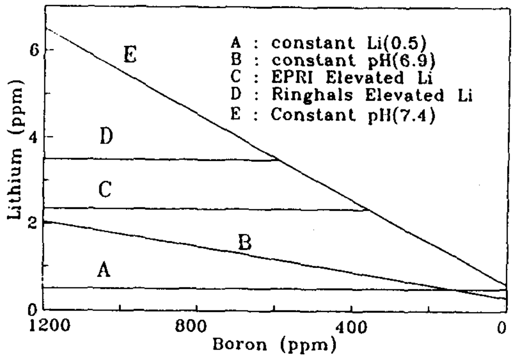

Fig. 7. The Assumed Changes of Lithium Concentration in KORI Unit 4 during Cycle 1.

modes. The actual chemistry data of the first cycle operation at KORI Unit 4 were utilized except lithium concentration. Mode A represents the constant Li of 0.5 ppm and mode B is the currently employed lithium-boron mode. Mode C represents the Elevated Li mode recently recommended by EPRI and mode D is also the Elevated Li mode, which has now being tested at Ringhal plants in Sweeden/17/. Mode E is the constant pH of 7.4 line which represent the zero temperature coefficients of iron solubility in nickel ferrites($Ni_{0.5}Fe_{2.5}O_4$).

The calculation results of activity buildup level

Talbe 4. Calculated Effect of Various Li Operation Modes on Activity and Iron Values in KORI Unit 4 during Cycle 1.

Operation Modes	Ex-Core			In-Core		
	Co^{58}	Co^{60}	Iron	Co^{58}	Co^{60}	Iron
Constant Li 0.5	3.553	0.337	0.071	15.94	0.861	0.055
Constant pH 6.9	3.158	0.298	0.075	10.99	0.566	0.044
EPRI Elevated Li	2.360	0.229	0.082	2.577	0.124	0.018
Ringhals Elevated Li	1.484	0.149	0.085	0.888	0.041	0.010
Constant pH 7.4	1.103	0.113	0.086	0.483	0.022	0.007

* Activity in $\mu Ci/cm^2$ and Iron in mg/cm^2

* Used actual chemistry values for KORI Unit 4 cycle 1.

are shown in Table 4. It is noticed in the table that the effect of pH levels on relative ex–core activity is quite big. Namely, if the pH increase 6.9 to the 7.4, the out–of–core activity level can be reduced by 65%. The reduction of in the in–core activity level is even more dramatic by increasing pH level. From our calculations, it is found that at higher pH (i.e., higher Li concentration), the corrosion products tend to deposit on out–of–core surfaces rather than on the fuel and that activity buildup of out–of–core can be reduced.

On the other hand, before application of the Elevated Li mode it is considered that the fuel integrity must be confirmed under the conditions of high pH operation mode, since the higher lithium concentration has a possibility of increasing fuel cladding corrosion/18/. But, if the nuclear fuel integrity is guaranteed by the vendor specification, it is recommended that pH should be as high as possible in a view of the great advantages for radioactivity management.

5. Conclusion

From the analysis of the water chemistry operation data in the operating domestic PWRs, it was confirmed in this study that most of domestic PWRs have been operated in accordance with the constant pH operation mode which is based on the solubility of magnetite and specified by the vendors.

Activity buildup levels were calculated for the two operation modes of Elevated Li chemistry and actual cheistry. The Elevated Li operation mode recently recommended by EPRI is based on the solubility of nickel ferrite which has recently been identified as a major component of corrosion products in the primary water system. The calculation results indicate that the Elevated Li operation mode could reduce the out–of–core activity buildup by about 25% compared with the actual chemistry mode during 1 cycle of plant operation. And if the plant is operated by Elevated Li mode through 3 fuel cycles (cycle 1,2 and 3), the out–of–core activity buildup can be reduced by about 35%. Furthermore, it is also shown that the out–of–core activities can be reduced by 65% during 1 cycle if the plant is operated at constant pH 7.4 instead of at pH 6.9.

In reality, the effect on the plant exposure rates might be a little different from the results shown in this study, since the calculation has been done assuming that only solutes in the primary coolant are responsible for the transport of radioactivity without consideration of the transport of the particulates and colloids which are also known to have a role in the crud transport process.

However, the results of this study suggest that increase of the coolant pH, or lithium concentration will significantly reduce the plant radiation levels judging from the application of the computer code. In addition, a plant test is recommended before application of the Elevated Li mode in the plant operation for examination of its operation effect on radiation exposure rates and the integrity of system structural materials and the fuel cladding.

Acknowledgements

The authors are grateful to C. B. Lee for his permission to use a solubility program in his dissertation.

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