Tracking Radioactivity Content in the Spent Fuel Pool in PWRs – 15150

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ABSTRACT

Over the last decade, significant progress has been made in managing radioisotopes generated in the PWR primary systems. These include limiting the use of cobalt containing materials, along with improved fuel performance and zinc injection to passivate the corrosion layers to limit the accumulation of Co-58 and Co-60 on primary coolant surfaces. Attention here is directed to the spent fuel pool where accumulation of spent fuel and activated material represents a growing source of mobile radioactivity supplemented with periodic exchanges of water from the reactor cooling system. Characterization of this source is increasingly important to effective management of disposal options. This paper examines the radioactive resin source term as determined from routine sampling of the coolant and fuel pool liquids. Targeted resin stream sampling and radiochemical analyses conducted at Diablo Canyon serve to inform and provide context for this examination. This examination is supplemented with computer modelling using the DW James Consulting computer program, 3R_SCAN.

At most operating plants daily reactor coolant samples are counted using gamma spectroscopy and recorded. These samples serve to track fuel performance and maintain water chemistry. A secondary use for this data is to maintain a day to day tracking of releases of radioisotopes important to waste disposal. Given the coolant concentrations, plant operating history, and corroborative mathematical models and sampling, accumulation of these radioisotopes in coolant cleanup system ion-exchangers can be predicted. Fuel pool sampling is less frequent, but at the same time, things change more slowly there. The fuel pool contains about ten times the volume of the reactor coolant but its cleanup system process rate is comparable to reactor coolant letdown, i.e. 5.047 – 8.833 liters per second [lps] (80-140 gallons per minute [gpm]). A spent fuel pool clean-up demineralizer flow rate is typically about 5.047 lps (80 gpm). Concentrations of longer lived radioisotopes in the spent fuel pool can be comparable to those in reactor coolant. Releases of corrosion products and fission products from the stored fuel occur at a low rate but given the overall inventory can be significant. Recognized scaling factor ratios, particularly those to Co-60, are skewed by the aging of the material in the pool. An additional consideration stems from refueling operations themselves. Prior to refueling, it is common practice to conduct a forced oxidation of the corrosion films within the reactor. This spikes the activity within the reactor coolant. Most of this activity is removed by the reactor coolant purification system prior to removing the reactor head. This paper examines the accumulation of activity in the fuel pool purification ion exchange resins and how it impacts the overall classification of those ion exchange resins. The following discussion focuses on the transport of nickel and cobalt isotopes particularly as it relates to determination of Ni-63 which has become increasingly prominent in disposal classification.

INTRODUCTION

No new plants have come on line since the mid-1980s. [1] Most of the current fleet of operating plants have accumulated more than 30 years of operation. Along with that they have accumulated more than 6 core loads of spent fuel. Fuel that may have been breached in the reactor will continue to release fission products and transuranic contamination on fuel assembly surfaces and exposed fuel in open pins. The rate of release is very slow. New production is insignificant but there is a lot of fuel. Recent radiochemical

sample results have shown an increased incidence of higher ratios between Ni-63 and Co-60 in mixed samples from spent fuel pool resins. Higher ratios signify that the cobalt 60 is depleted from production ratios relative to Ni-63 and other longer lived isotopes. The extent to which the spent fuel pool is becoming a more prominent contributor to this source term is examined in this paper.

The Fuel Pool By Design

Fuel pools are designed to be free of penetrations below a level that would result in uncovering the stored fuel. The lowest penetration is located to ensure sufficient coverage of water above the fuel to enable access to the refueling floor. Fuel Pool parameters as discussed in this paper are drawn from the Byron UFSAR, Section 9. [2] Water to the fuel pool cooling and cleanup system is drawn from a penetration 6-8 feet below the upper surface of the water and returned through a penetration at about the same elevation on the opposite side of the pool. The return line is extended downward to about 6 feet above the fuel racks. Heat from the stored fuel is carried by convection currents into a mixing zone between the supply and return elevations.

Release of Activity

The basic source of activity in the spent fuel pool is production from stored fuel. There are effectively no new radionuclides being produced in the stored fuel. Releases to the spent fuel pool from stored fuel are composed of radioisotopes that were there when the fuel was deposited in the pool. Based on post outage fuel pool activity measurements, it appears that elevated levels of contamination are present on fuel assemblies as they are brought in for storage. Transport from the fuel surfaces appears to be driven by higher temperature levels in the fresh fuel which accelerates release from the fuel. Once the fuel is placed in storage, elevated release rates continue as the fuel starts to cool down. Following each refueling the new fuel tends to dominate the release rates throughout the cycle until the next refueling occurs.

There is typically a five day lag time between the start of shutdown and forced oxidation until the plant is ready for fuel transfers. By this point, the radionuclides concentrations are back in equilibrium with the letdown cleanup systems. Transfers of activity between the RCS and fuel pool through exchanges of water should have minimum impact. The activity is transferred on the fuel assembly itself and through various surface interactions with water in the fuel pool and is released at levels that roughly trail the cool down of the decay heat.

Both Diablo Canyon and Byron are two unit Westinghouse PWRs. [1] Diablo Canyon has separate fuel pools for each unit while Byron shares its pool between the two units. Both plants perform staggered refueling on an 18 month cycle with a 1/3 core replacement at each refueling. Both plants were brought into service in the mid-1980's and are approaching 30 years of service. Diablo Canyon completed the 18 refueling cycle on both units in 2014. Over that time span each unit has contributed about 5 equivalent cores of spent fuel. With that rate of transfer there are continuing strong transients of temperature in the spent fuel pool. Figure 1 below shows heat loadings to the Byron spent fuel pool with the buildup of spent fuel.

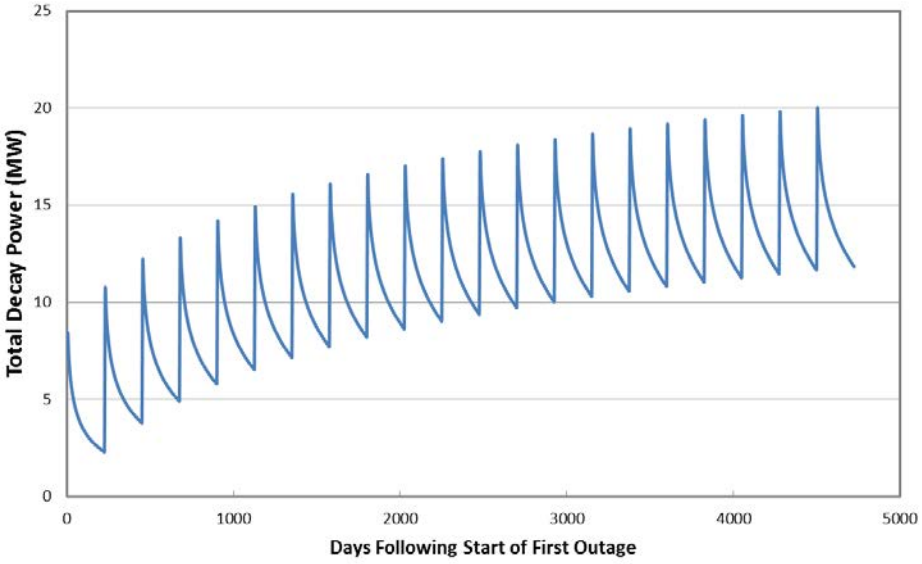

Figure 1 Decay Heat Cycles Following Refueling

In this case, the heat generation is calculated according to the following equation: [3]

$$\frac{P}{P_0} = 6.1\ x\ 10^{-3}(\tau - T_0)^{-0.2} - \tau^{-0.2}\ [3] \qquad \text{(Eq. 1)}$$

Where:

P = current power (decay heat)

P_0 = operating power

τ= time from start of irradiation to present

T_0 = total irradiation time

The basis for the heat generation is a dual unit pressurized water facility with shared fuel storage (eg, Byron). Each plant operates at 3500 megawatts thermal. As can be seen in the graph, the thermal loading from each off-load drops rapidly to about 2.3 MWt over the duration between cycles. While the decay heat from each cycle continues to drop going forward, the cumulative effect raises the heat input to around 8 MWt at the low point of the 21st off-load. There is certainly enough heat introduced into the pool to create strong convection currents and to maintain substantial mixing from the lower levels of the pool. The pool itself is designed to maintain water temperatures during refueling to below 60 degrees C (140 degrees F). Given the levels of heat generation during early weeks of storage the fuel pool is effectively a cauldron.

Activity Transfer

As fuel is off-loaded from the reactor, moved through the transfer canal, and across the fuel pool it's widely speculated that each assembly leaves a "trail" of activity. This doesn't provide an explanation for the entire process. If activity was only introduced during transfer it would be quickly cleaned out by the pool cleanup system. Figure 2 shows the Cobalt 58 concentration measured at the inlet of the fuel pool demineralizer. Clearly evidenced from the Co-58 tracked at the Byron Station, there is continuing release at levels high enough to maintain elevated Co-58 activity long after the end of the refueling.

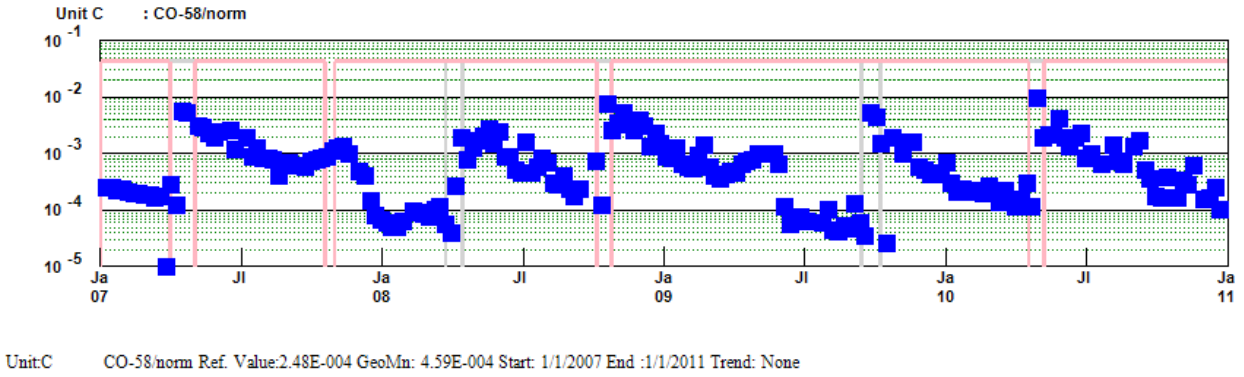

Figure 2 Co-58 Measured Fuel Pool Activity Concentrations

Figure 2 demonstrates a very clear impact of activity migration stemming from fuel transfer operations. It is strongly demonstrated in the case of Co-58 since what is already in the pool has decayed by at least a factor 8 before the next outage is initiated. Overall the activity of Co-58 (assuming the pool is well mixed) increased by an order of magnitude. In the case examined, almost 3.7E+11 Bq (10 Ci) of Co-58 activity were transferred in the first cycle shown.

To supplement this discussion, the following parameters are adopted;

Table I. Fuel Pool Parameters

Parameter	**Value**	**Units**
Pool Volume	61000	ft^3
	1726300000	cm^3
Pool Cleanup Rate	80	gpm
	5046.7	cm^3/sec
Cleanup Coefficient	2.92E-06	sec-1
Lambda Co-60	4.17E-09	sec-1
Lambda Ni-63	2.20E-10	sec-1
Lambda Co-58	1.13E-07	sec-1
Concentration Co-60	1.00E-03	µCi/cm^3
Concentration Co-60	37	Bq/ cm^3
Concentration Ni-63	1.00E-03	µCi/cm^3
Concentration Ni-63	37	Bq/ cm^3
Co-58 (Directly Following Outage)	5.70E-03	µCi/cm^3
Co-58 (Directly Following Outage)	2.11E+02	Bq/ cm^3
Co-58 (225 Days Later)	0.00029	µCi/c m^3

Parameter	Value	Units
Co-58 (225 Days Later)	1.073E+01	Bq/ cm^3
Total Co-58 Initially Transferred	9.84	Ci
Total Co-58 Initially Transferred	3.641E+11	Bq
Decay Removal	9224915.625	270 days
Co-58/Co60	25	average of two units over 4 year time span
Co60 addition	0.3935964	

The basic formulation of the transport is represented by the following equation:

$$\frac{dC(t)}{dt} = Release(t) - C(t)(\beta + \lambda) \qquad \text{(Eq. 2)}$$

Where:

C(t) = Concentration as a function of time *t*

Release(t) = time dependent release from fuel surfaces

β = Cleanup removal constant (sec^{-1})

λ = Decay Removal Constant (sec^{-1})

The cobalt 58 trace is used to establish general parameters for the transfer of activity during refueling. Fuel pool radiochemical samples are collected on a weekly basis, so there is some uncertainty as to how accurate the starting point is reflected.

In any case, what we observe is a spike in pool activity inventory of almost 3.7E+11 Bq (10 Ci) following the outage. This spike gradually dissipates over time to the next outage – figured to be about 250 days. Considering the half-life of Co-58, a total of 3.52 half-lives pass during the 250 days resulting in a decrease from the initial amount by a factor of 11.5. Considering that the cleanup system has been operating during the entire period, a continuing source of activity has been introduced that wasn't there before. If we assume that the cleanup demineralizer functions at about 95 percent efficiency for cobalt we would see removal of about 300 curies of additional Co-58 activity during the interim period between outages. To complete the activity balance, it's assumed that additional Co-58 activity is introduced into spent fuel pool at an average rate of 5.55E-01 Bq/sec (1.5E-05 µCi/sec). It starts at a higher rate and ends lower in accordance with decay off of the source term accompanying reduction of decay heat. At this point the fuel is well settled in the racks, but still basically contributes the continuing activity release. It is speculated here that the high decay energy in the off-loaded fuel is creating conditions favorable to release of surface activity and generation of convection gradients to carry the activity up from the fuel racks. What we have to support this belief is that the activity wasn't in the fuel pool before the refueling and there was no comparable generation of activity occurring. The elevated release had to be brought in with the fuel. Basically, all of the activity dealt with here is accounted for above the equilibrium or average activity content of the fuel pool.

Normal practice in preparing the reactor for the outage to allow the letdown clean-up system to normalize activity concentrations in reactor coolant prior to the start of refueling operation. It is not particularly likely that the excess activity is transferred through the coolant. Figure 3 below provides a calculated Co-58 inventory in the spent fuel pool following a typical concentration transient. Again, nearly all of the activity reflected below was introduced from the preceding refueling.

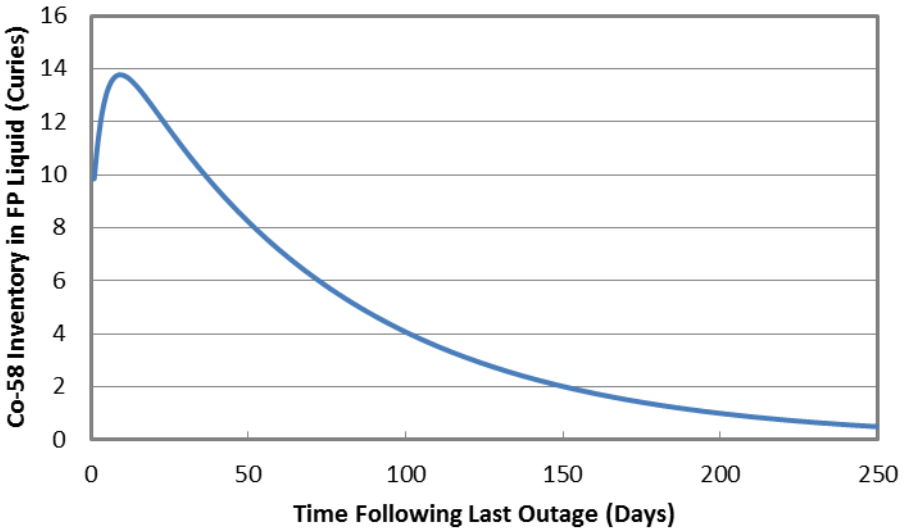

Figure 3 Co-58 Activity Inventory in Spent Fuel Pool

Transport of Co-60

The demonstration is not so apparent in the case of Co-60. As can be seen in the Figure 4, the outage spikes of Co-60 are not as apparent as those of Co-58. This again appears largely due to the concentration of Co-60 in the reactor coolant system being closer to the fuel pool concentration. Never-the-less there are bumps in activity of Co-60 along with the Co-58 and these are observable and expected to follow a comparable transport path to that of Co-58.

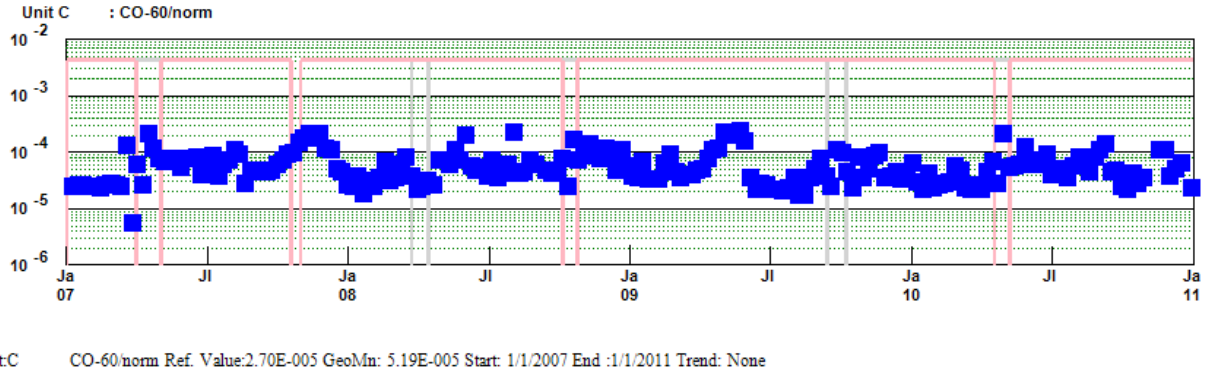

Figure 4 Co-60 Measured Fuel Pool Activity Concentrations

Figure 5 shows reactor coolant cobalt traces for units 1 and 2 of the Byron plant. Co-60 concentrations in the reactor coolant are estimated from a geometric mean of 1.46E+00 Bq/cc (3.95E-5 µCi/cc) for unit 1 and

3.37E+00 Bq/cc (9.12E-5 µCi/cc) for Unit 2. The fuel pool concentration during roughly the same interval averages 2.20E+00 (5.95 E-5 µCi/cc) basically and is enveloped by the coolant concentrations. As we can see with the coolant traces is that the value from day to day is somewhere between 1E-4 and 1E-5. Given the large number of samples and the central tendency, we can determine the average value from this data with a high degree of confidence. Even accounting for some spiking during the forced oxidation step, the values, as noted above, have mostly settled down by the start of fuel transfer.

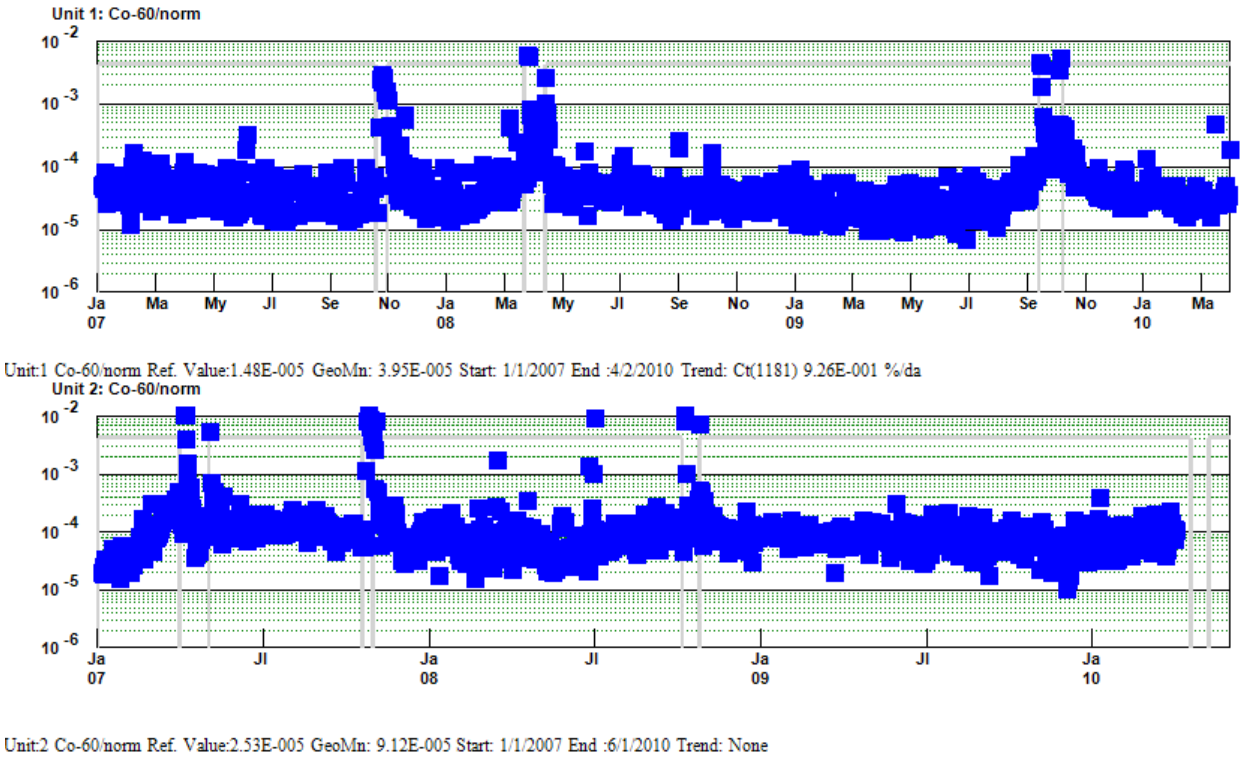

Figure 5 Co-60 Measured Activity Concentrations in Reactor Coolant (Two Unit Facility)

The transport and fuel pool burden of Co-60 can be estimated following the same process we have developed for Co-58. The initial concentration of Co-60 was estimated from Figure 5 to be about 7.4E+00 Bq/cc (2.0E-4 µCi/cc); from the same data it is assumed that the final activity is reduced by about a factor of 10. With a cobalt concentration of 7.4E+00 Bq/cc (2E-4 µCi/cc), multiplying this by the coolant volume the total Co-60 mobile inventory is about 1.28E+10 Bq (0.345 Ci). Note that we haven't assumed that there is anything else in the pool. So accounting for decay and cleanup removal, an additional 1.17E+11 Bq (3.17 curies) had to be transferred during the refueling operations. As in the case of Co-58, Co-60 continues to be released at elevated levels.

Transport of Ni-63

There is no direct trace in gamma spectrum data for Ni-63. However, we can figure out how to determine Ni-63 from sample data collected by Diablo Canyon (DCPP). DCPP has the ability to sample individual resin streams during transfer into their spent resin storage tank. For the past few years DCPP has collected a series of samples to track changes in isotopic ratios. The changes are due to the replacement of large components (eg, steam generators and reactor pressure vessel heads) and are brought about by zinc injection into the primary coolant to more effectively block cobalt depositions in the reactor coolant system surfaces. Ratios calculated from the DCPP samples from 2011, 2012 and 2013 are plotted in Figure 6 below.

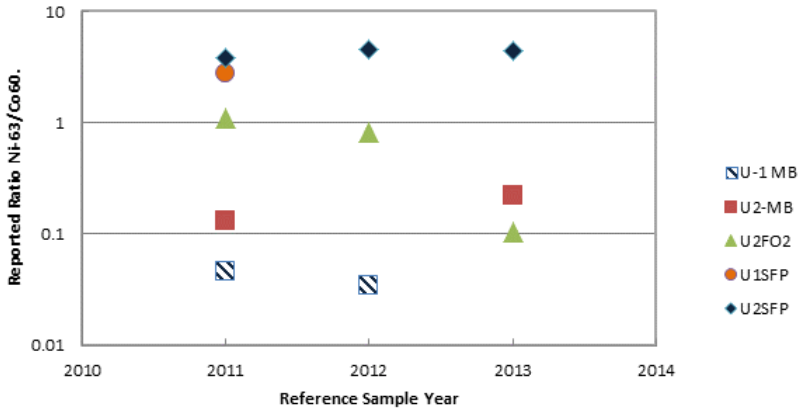

Figure 6 10CFR61 Sample Results for Ni-63/Co-60 (DCPP Experience)

Streams displayed include Units 1 and 2 CVCS letdown mixed beds, Unit 2 CVCS forced oxidation shutdown cleanup resin beds and Units 1 and 2, spent fuel pool beds. Some observations made in the data are the mixed bed ratios reflecting removal during normal operations are exhibiting ratios well below 1. The Unit 2 forced oxidation bed shows a decrease in the Ni-63/Co-60 ratio of about a factor of 10 over 3 cycles. The fuel pool ratios are elevated for both units with Unit 2 at a relatively steady ratio between 3 and 4. Interestingly, the mixed bed ratios and forced oxidation bed ratios are within the ranges of production ratios for materials common in PWRs. This is contrary to an expectation that ratios are increasing within the RCS, rather it confirms that ratios are increasing in the fuel pool. Both Byron and Diablo Canyon have operated for a period of about 10,000 days (25-30 years). For the purpose of the following discussion, it will be assumed that the Ni-63/Co-60 production ratio is about 0.5. This would be reflective of Inconel type materials and consistent with industry experience in general.

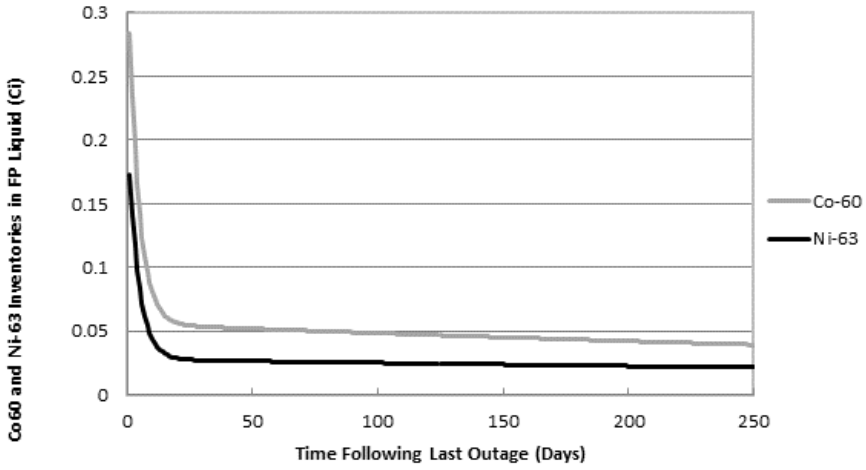

Figure 7 Co-60 and Ni-63 Activity Inventory in Spent Fuel Pool

Projected Ni-63 and Co60 activities in the spent fuel pool are shown in Figure 7. In this case it is assumed

that the activity from each transfer is decayed from the time it was deposited. As we have already seen above, the dominant source of activity can be attributed to the most recent transfer which temporarily overwhelms the pool inventory. The expectation would be that the nickel cobalt ratio will reflect the lower ratio associated with the most recent transfer. The average ratio projected around 10000 days corresponds relatively closely with the ratio observed at DCPP for fuel pool resins.

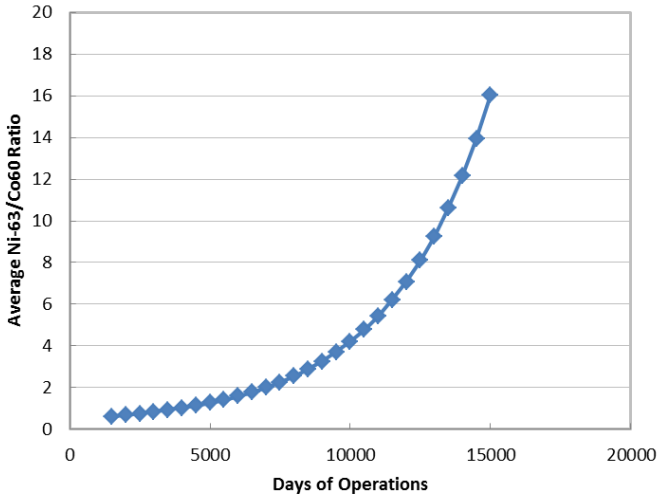

Figure 8 Projected Average Ni-63/Co-60 Ratio in Spent Fuel Pool

There is one more bit of tempering that is necessary to arrive at a final conclusion. As speculated above, the release of radioactivity appears to be temperature driven. Given the high levels of mixing and the large amounts of activity brought in with the most recent fuel off load we would expect the mixed ratios to be dampened by the lower ratios in the freshest material. Figure 8 shows a rolling average based on each step having an equal probability of release. At best this is a conservative view, but not entirely inconsistent with recent sampling experience.

CONCLUSION

Characterization of radioactive waste is fundamentally important to meeting the moral and regulatory obligations for waste disposal. It should be approached with a broad understanding of where, when and how the waste was generated, what is the physical nature of the waste, and what is the source of radioactivity in it. This paper seeks to resolve how mobile radioactivity collects and behaves in spent fuel pools and how it impacts ultimate disposal of cleanup resin from the spent fuel pool. It provides an explanation as to how elevated Ni-63/Co-60 ratios occur in the pool and projects into future observations. Command of this information enables the operators to develop the process knowledge and context to more effectively managing the inventories of activity generated in process waste streams.

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