ACTIVITY BALANCE IN ONTARIO HYDRO PRIMARY HEAT TRANSPORT SYSTEM AS INFORMATION FOR RADIOACTIVE WASTE MANAGEMENT

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ABSTRACT

This Radioactivity Balance has been performed as a demonstration study on the Primary Heat Transport (PHT) system of selected Units of Ontario Hydro's Pickering, Bruce A and Darlington nuclear stations. PHT sample radiochemistry analysis, radiation field surveys, analysis of deposits, Gaseous Fission Product on-line monitor (GFP) data and CRUD analysis each provides different information on radioactivity in the PHT system.

The unique CANDU reactor technology, with heavy water coolant limits the applicability of experiences of others and enhances the need for developing a CANDU specific activity transport-monitoring program. This paper describes some experiences gained from CANDU PHT system activity transport monitoring and how this information can be used in Radioactive Waste Management and in decommissioning planning.

Activity balance and activity transport information is useful for several purposes. Radiochemistry analysis and GFP on-line monitoring provide information about PHT chemistry and fuel integrity. Different sampling locations can provide information on removal efficiencies and removal rates of radionuclides from the PHT. The radioactivity of filters and spent resins can be calculated for waste management purposes. Radiation field surveys provide information about activity distribution, growth and composition within the PHT system. This information can be used in dose exposure, maintenance and decontamination planning. Deposit analysis results provide detailed information on activity deposition in steam generators and can be used for Co-60 deposition rate calculations.

In this paper, an evaluation and comparison of data collection methods as well as an integral approach to different sources of information, is presented. Distribution of radioactivity in the PHT system and its input into Waste Management Systems is discussed.

In PHT heavy water samples, besides short-lived I-131 and gaseous fission products, the highest contributions to total radioactivity is from shorter lived radionuclides. The contribution of Sb-124 and Zr-95 is higher than that of Co-60. Radiation field surveys show that the dominant contributors to radiation fields are Co-60 and Sb-124. The Co-60 contribution is dominant and is higher in boilers than in the rest of the PHT system. The Co-60 contribution to radiation fields increases with the length of unit operation. The deposit analysis results have shown that the dominant deposited radionuclide is Fe-55, which emits x-rays and does not pose a significant radiation hazard. Deposit analysis by the Oxiprobe method has shown that significant quantities of alpha emitters are present in the steam generator deposits. Chemical speciation, Oxiprobe data

and decontamination factors can be used to estimate the distribution of radioactivity on filters, ion exchange resins and on the primary side steam generators. From the perspective of radioactive waste storage and disposal, only radionuclides with long half-life such as Co-60, Cs-137 and longer are significant. The best information about those radionuclides is obtained from Oxiprobe measurements.

INTRODUCTION

In 1996 and 1997, activity balance studies of the PHT system were performed for Bruce A, Unit 3 (Ref. 1), Pickering Unit 1 (Ref. 2) and Darlington Unit 2 (Ref. 3). The findings of these studies are summarized in this paper, with an additional discussion of the applicability of the information to Radioactive Waste Management.

The purpose of the studies was to provide a baseline for activity transport monitoring, to perform activity balance calculations, and to identify any trends in concentration and distribution of monitored radionuclides. The studies also compared the generation rate of specific radionuclides with measured radionuclide activity within the Primary Heat Transport (PHT) system. The activity transport analysis provides information about the short and long term distribution of radioactivity in the PHT system. Furthermore, the objective was to develop recommendations for a comprehensive data collection protocol to generate a valid database for a continuous activity transport monitoring program.

The radioactivity present in the PHT system is generated in the core of the reactor from two main sources. The first source is fission, which produces a wide spectrum of radionuclides. The second source is activation of dissolved and suspended materials. The majority of these activated materials are corrosion products.

In both cases, the activity may be transported out of the reactor core in solution or as a suspended solid. Outside the reactor core the radionuclides may remain in the heavy water or may deposit on PHT system surfaces. Radionuclides can be removed from the PHT system by the purification system, through D_2O leakage or through spent fuel removal. Radionuclides deposited in the PHT system may be also removed by system decontamination.

The relevant operating parameters for nuclear power units are given in the first section of this paper. In the second section, sampling and radiochemistry analysis methods and radiochemistry results are evaluated. The CRUD analysis data are also discussed in this section. The third section describes obtained by radiation field surveys results. The fourth section presents the Oxiprobe results. In the fifth section the Gaseous Fission Monitoring (GFP) results are analyzed. Comparison of data from different sources and calculated activities is discussed in the sixth section.

Relevant Plant Operational Parameters

Table 1 data lists the relevant operating data in the observation periods. The observation period of time was between 3 months (Bruce A, Unit 3) and 12 months (Darlington, Unit 2).

Table I Relevant Operating Data

Parameter	Pickering, Unit 1	Bruce A, Unit 3	Darlington, Unit 2
Time period	June 9,1995 – April 15,1995	January 1,1996-May5,1996	Jan. 1,1996-Dec.31 1996
D_2O in PHT system	154.96 m^3	279.7 m^3	217.2 m^3
Energy Produced	11585 GWh	6422 GWh	21451 GWh
Heavy water Loss	5.8 kg/h	10 kg/h	64 000 kg
Non recovered loss	N/A	N/A	1914 kg
Average purification flow rate	12 600 kg/h	28 188 kg/h	34 560 kg/h
Thermal power	1744 MW	2832 MW	2776 MW

DATA INFORMATION SOURCES FOR ACTIVITY TRANSPORT STUDY

In the Activity Transport studies, different sources of PHT system radioactivity distribution information, were used to balance radioactivity in the system. Possible changes in data collection methods to improve Activity Transport monitoring, were evaluated.

PHT System Sampling and Radiochemical Analysis

Historically, the primary reasons for radiochemistry analysis of PHT system samples were to monitor iodine production to comply with health and safety targets in case of an accident and to track the production of other radionuclides. Sampling points and sampling programs were not designed to monitor radioactivity transport throughout the system. For example, purification inlet and outlet samples were monitored only occasionally at the Darlington station.

Typically, PHT system samples are taken at the north and south reactor outlet header. Aqueous samples from the PHT system are then analyzed by gamma spectrometry.

For this activity transport study, the radionuclides tracked in the PHT system were selected based upon their detection frequency as well as their half-life and health risk. The selected radionuclides for Darlington were: Ce-144, Cs-137, Co-60, I-131, Fe-59, Nb-95, Sb-124, Sn-113, and Zr-95. In order to be selected, the half-life of the radionuclide or of its daughter has to be long enough to be detectable some time after shutdown. The minimum half-life was arbitrarily selected to be 15 d. The selected list of radionuclides at Bruce and Pickering NGS also included: Ce-139, Cr-51, Cs-134, La-140, Mn-54, Rb-89, Sr-89 and Zn-65. These radionuclides were not detected in the Darlington PHT system during the monitoring period.

There are also some radionuclides that should be tracked that are not gamma emitters but are of interest because of their impact on the environment, health and safety issues or waste management. These radionuclides are H-3, C-14 and Sr-90. The first two are determined by liquid scintillation and chemical separation. Beta counting is used to determine Sr-90. Typically there is less data for these radionuclides.

The number of radionuclides tracked in this study is significantly smaller than the detailed lists of theoretically calculated core equilibrium activities of radionuclides. The majority of radionuclides from the theoretical list cannot be detected easily in the PHT system [References 1,2 and 3].

The activity trends for Co-60 and Cs-137 were monitored. Based on the raw data, it appears that Co-60 has a reasonably stable concentration with occasional spikes. The largest spike occurred in the shutdown period. The Cs-137 concentration oscillations seem to be somewhat larger, but stabilise towards the end of the study period. The average measured concentrations of Co-60 and Cs-137 used in the activity balance are shown in Table 2.

Table II: Average Measured Concentrations of Cs-137 and Co-60 at Studied Stations

Radionuclide	**Bruce A**	**Pickering**	**Darlington**
Cs-137 (operation)	0.007 (μCi/kg)	0.003 (μCi/kg)	0.4 (μCi/kg)
Co-60 (operation)	0.022 (μCi/kg)	0.73 (μCi/kg)	0.93 (μCi/kg)

The activity concentration data was back calculated to represent the concentrations present at the actual sampling time. Bruce and Pickering also perform gamma counting a week after sampling to allow short lived radionuclides to decay.

There was significantly less gamma spectrometry analytical data available for the shutdown periods. However the study at BAND [Reference 1] showed that there was a significant difference between the concentration of specific radionuclides in samples taken during operation, and those taken during shutdown. As well, a number of radionuclides were detected during shutdown that were not detected during operation. The measured activity of Cs-137 during shutdown was 28 times larger than measured during operation. The measured activity of Co-60 was four times higher during shutdown than during operation. Additional study is required to explain the difference between the samples taken during operation and those taken during shutdown.

In Table 3, the average radionuclide concentrations from Darlington CRUD data are presented. The averages are from only two CRUD measurements. The results are expressed as concentration of radionuclides per kg of the PHT coolant. As expected Cs-137 was not detected on the CRUD samples. The concentration of Co-60 is significantly lower than measured in liquid samples. Therefore, a part of Co-60 is probably, either in the dissolved form, or more likely in the form of colloidal particles that pass through the CRUD filter.

More sets of data would be required to provide a more detailed comparison of CRUD results with concentrations in the liquid PHT sample.

Table III Average Radionuclide Concentrations in CRUD Samples

Radionuclide	Average Concentration in PHT (μCi/kg)
Sb-124	3.2E-03
Ce-141	1.1E-04
Ce-144	3.7E-04
Cs-134	2.1E-03
Co-60	2.0E-04
Hf-181	9.3E-05
Ho-166	6.3E-01
La-140	7.6E-1
Nb-95	1.0E-03
Ru-103	1.2E-03
Ru-106	6.4E-04
Tb-158	1E-04
Sn-113	1.1E-04
Zr-95	1.5E-03

Chemical Speciation of Radionuclides

The objective of chemical speciation is to determine the chemical species or chemical form of the radionuclides detected in the PHT. The speciation data is useful information for activity transport monitoring and the activity balance. Chemical speciation can be used for predicting the distribution of radioactivity in the PHT.

Speciation data was available for Bruce A, Unit 1, because a method to determine speciation of radionuclides was developed using a PHT sample originally collected to calibrate an iodine cartridge. A one-liter HTS sample was passed through a series of filters starting with a Millipore filter on top, followed by a cation paper and then two anion papers. The PHT sample was counted on a γ- spectrometry system before filtration and after filtration through each consecutive filter. All filters were also counted on the γ- spectrometry system. From the analytical results, the probable chemical form of the radionuclides was determined and is presented in Table 4.

The validity of these speciation results are questionable because of the sampling techniques used at Bruce A, and it is not known whether the results at Bruce A can be applied to other stations. It is likely that some insoluble species may have deposited in sampling lines. As well, because the analysis did not take place under operating conditions, there may have been a change in solubility of some radionuclides due to cooling and depressurization of the HTS sample. The results for Co-60 speciation at Bruce A are different than what was found in CRUD analysis at Darlington.

At Darlington Co-60 was not removed by CRUD filter. This can be explained with different filter pore (mesh) sizes, but there may be other factors as well. Within the PHT, insoluble radionuclides will likely be removed by the PHT purification system filters or will deposit on system surfaces such as boiler tubes. The soluble chemical forms will most likely be removed by IX resins.

Table IV Chemical Speciation at Bruce A PHT System

Radionuclide	Probable Form	Origin	Probable State in Solution
Ba-140	Ba^{2+} salt	Fission product	suspended
Ce-144	Ce^{3+} and $Ce(OH)_3$	Fission product	partly dissolved and partly suspended.
Co-60	$Co(OH)_2$ or CoOOH	Neutron activation Co-59	suspended particle
Cs-134	Cation salt	Fission product	dissolved
Cs-137	Cation salt	Fission product	dissolved
I-131	I^-, IO^-_x	Fission product	dissolved
I-133	I^-, IO^-_x	Fission product	dissolved
I-135	I^-, IO^-_x	Fission product	dissolved
Kr-85m	Kr	Fission product	entrained gas
La-140	La^{3+} and probably $La(OH)_3$ or other form.	Fission product	partly dissolved and partly finely suspended solid.
Mn-54	$Mn(H_20)_6^{2+}$	Fe-54(n,p), Mn-54	stable dissolved species
Mo-99	MoO_4^{2-}	Fission product	dissolved
Na-24	Na^+	Neutron activation Na-23	dissolved
Nb-95	Halides possibly	Fission product and neutron activation product.	suspended
Sb-124	SbO+ salt, possibly other form.	Fission product and neutron activation product.	suspended
W-187	WO_4^{2-} most probably.	Neutron activation of W-186	dissolved
Xe-131m	Xe	Fission product	entrained gas
Xe-133m	Xe	Fission product	entrained gas
Xe-133	Xe	Fission product	entrained gas
Xe-135	Xe	Fission product	entrained gas
Zr-95	Zr^{4+} salt	Fission and neutron activation product.	suspended

Radiation Field Surveys

Radiation field surveys are performed by Chemistry Department personnel to measure the average γ- fields for the reactor faces and steam generators (boilers), as well as radionuclide contributions to total γ- fields. The radiation fields are calculated to ten days after shutdown so

that historical data are comparable. The status of the equipment, such as whether the boilers are full or empty, are used to equalise the survey data.
The results of radionuclide contributions to radiation fields from typical radiation field surveys of Pickering and Bruce Units performed before the Activity Balance Studies are presented in the Table 5.

The steam generator Co-60 radiation fields versus the Equivalent Full Power Days (EFPD) are presented in Figure 1 for Bruce A. The averages of the percent contribution were calculated by averaging the percent contribution to the field of each radionuclide at each survey location. By merely taking the average of these data, it is assumed that the total activity measured at each of the survey locations was approximately equal and hence the data are comparable. This may not be the case. Also, if a radionuclide is below the Minimum Detectable Activity (MDA) at one or more locations, it lowers the average significantly.

The reactor survey results reveal that Co-60 and Sb-124 are the largest contributors to the reactor fields, followed by La-140. The percent contribution of Co-60 to boiler radiation fields presented in the Table 5 is typical for reactors with high Effective Full Power Days (EFPD) of operation (3000 - 4000).

Table V: Typical Radionuclide Contribution to Radiation Fields at Bruce A and Pickering

Radionuclide	Half-life	Average of % Contribution to the Field, Bruce A	Average of % Contribution to the Field, Pickering	Average of % Contribution to the Field, Bruce A	Average of % Contribution to the Field, Pickering
		Reactor Face		Boilers	
Cs-134	2.065 y	ND *	0.15	ND	0.08
Cs-137	30.17 y	1.3	0.2	0.2	0.3
Co-60	5.27 y	61.5	38.8	76.9	70.0
Fe-59	44.5 d	4.3	4.5	1.9	2.4
La-140	12.75 d (Ba-140)	ND	19.5	ND	6.2
Mn-54	312.2d	0.6	0.4	0.05	0.18
Nb-95	35 d	6	4.25	3.3	1.9
Ru-103	39.3 d	0.08	0.65	ND	0.03
Ru-106	1.02 y	0.6	0.2	1.1	0.3
Sb-124	60.2 d	23.2	29.6	15.3	17.7
Zn-65	243.8 d	ND	0.05	ND	0.008
Zr-95	64 d	2.5	1.75	1.4	0.7
* means that the concentration is below MDA					

Some general conclusions, based on the trend observations from several radiation fields survey reports [Reference 4], can be made. Typically the total γ-fields at both the reactor face and the boiler tend to plateau after a significant number of EFPDs have elapsed. This implies that there is no net increase or decrease in γ- fields. The EFPD required to reach this steady state is usually > 2000 days depending upon the Ontario Hydro unit. Upon reaching this steady state, the radiation fields tend to fluctuate with a range of a few mR/h. Once steady state has been achieved in terms of activity, the contribution of Co-60 to radiation fields becomes even more significant (75-95% as compared to 50-80 % at the beginning of station operation). This steady state pattern can be disrupted by a CAN-DECON (full system decontamination), or fuel defects.

Results shown in Figure 1 were used to estimate the increase of the Co-60 steam generator radiation fields with EFPD for Bruce A, Unit 3. The equation for the best-fit straight line in Figure 1 to determine of the Co-60 radiation field is:

$$D = 0.0018(\text{EFPD}) + 0.3 \quad \ldots..(1)$$

The slope from this equation (0.0018) was applied to determine the Co-60 deposited in boilers at Bruce during the study period.

A similar relationship was determined for PNGS Unit 1. Radiation field survey data after the unit retube was used to determine the slope. The rate of radiation field increase was steeper in this case. This may be explained by the Co-60 inventory that remained in the PHT system after the retube.

OXIPROBE SAMPLING AND ANALYSIS

Oxiprobe is a testing method, that enables identification and quantification of radionuclides in deposits on sections of the primary side of steam generator tubing. Oxiprobe measurement has been done at BAND, Unit 3 and at PNGS, Unit 1, however it has not been done at any DNGS unit.

The analysis gives information about the mass and radionuclide content of the deposits on steam generator tubes. Deposition rates can be calculated, if correlated with operational history.

Oxiprobe involves dissolving all of the deposits in the section of the steam generator tube where the probe is placed. The solution is then analyzed by Inductive Plasma Spectroscopy (ICP) for metal ion concentrations, and by gamma spectrometry for gamma emitting radionuclides. Samples are also usually analyzed for alpha and beta activity. Results from Oxiprobe sampling and analysis are a useful source of information in activity transport studies [References 4 and 5].

Oxiprobe results, shown in Table 6, have revealed that the most abundant radionuclide in steam generator primary deposits is Fe-55. Fe-55 is not detected during radiation field surveys or PHT system sample γ- spectrometry, because it is not a γ emitter, but emits only x rays that follow an electron capture transformation. Oxiprobe results also showed that there was a significant quantity of α emitters in the deposits (up to about 200 Ci in total for BAND Unit 3 boilers).

This information is particularly significant from the waste management and decommissioning perspective because it may be a major factor in waste characterization and categorization.

ON-LINE RADIOACTIVITY MONITORING AT DNGS (GASEOUS FISSION PRODUCT MONITORING)

The purpose of the PHT Radioactivity Monitoring System (also known as the Gaseous Fission Product Monitoring System -GFP)is to detect failed fuel at an early stage in the failure process. The system is in full time use only at the Darlington station.

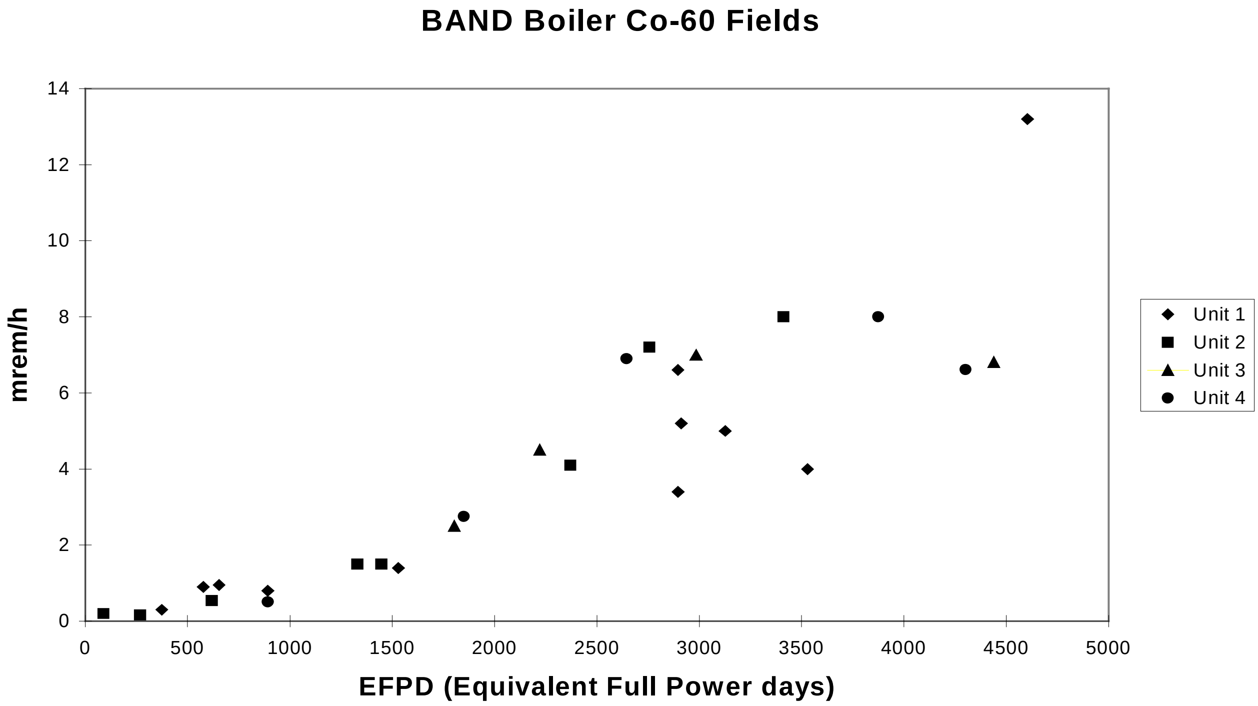

Fig. 1. Co-60 radiation field trends at Bruce A Units

When fuel cladding fails, radio -iodines and other fission gases as well as some activated radionuclides are released into the PHT coolant. Gaseous fission products released into the coolant through the defective cladding would lead to an increase of fission product activity concentration in the coolant. This increased activity of fission products in the coolant could constitute a hazard. Early detection of failed fuel is required, so that the operator can undertake appropriate action. The focus is usually placed on observing the concentration increase of the following radionuclides: Xe-133 (81 keV γ ray), Kr-889 (196 keV γ ray), Xe-135 (259 keV γ ray) and I-131 (364 keV γ ray). However, it was also realized that the system could be used to monitor concentration of other radionuclides including major activation products.

Table VI Radiochemistry and Chemistry Data from Oxiprobe Measurements at PNGS, and Bruce A

	Average Concentration (µCi/g of deposit)	Estimated Activity (Ci)	Average Concentration (µCi/g of deposit)	Estimated Activity (Ci)
Radionuclide	Bruce A (Unit 3)		Pickering (Unit 1)	
Ag-110M	0.052	0.13	0.041	0.080
Am-241	0.069	0.16	1.40	2.73
Ba-140			0.30	0.58
Ce-141	0.52	1.25	5.16	10.06
Ce-144	3.5	8.4	72.38	141.14
Co-58	0.003	0.007	0.098	0.19
Co-60	29.8	72.1	22.13	43.15
Cr-51	0.094	0.23	0.45	0.88
Cs-134	0.005	0.01	0.23	0.45
Cs-137	0.01	0.24	1.27	2.48
Eu-152	0.008	0.19	0.02	0.039
Eu-154	0.16	0.38	3.46	6.75
Eu-155	0.19	0.46	2.85	5.56
Fe-59	0.68	1.65	2.00	3.9
Hf-181	0.24	0.058		
La-140	0.0034	0.009	0.34	0.66
Mn-54	0.84	2.0	1.61	3.14
Nb-94	0.05	0.13	5.71	11.13
Nb-95	4.15	10		
Ru-103	2.04	5.3	8.38	16.34
Ru-106	7.05	17.1	66.75	130.16
Sb-124	2.38	5.8	8.5	16.58
Sb-125	0.39	0.95	3.25	6.34
Sn-113	0.11	0.27	0.48	0.94
Zn-65	0.37	0.9	0.055	0.11
Zr-95	2.3	5.6	2.87	5.60
Fe-55 (X-ray emission after electron capture)	50.8	122.9	215.67	420.56
Sr-90 (β-decay)	0.094	0.23	37.17	72.48
Pu239+240 (α-decay)	11.87	28.7	1.77	3.45
Pu-238 (α-decay)	6.15	14.9	0.68	1.33
Am-241 (α-decay)	29.4	71.1	1.51	2.94
Cm-242 (α-decay)			0.077	0.15
Cm-244/245 (α-decay)	92.8	224.5	0.59	1.15

Note:1)
Range of concentrations taken from the results of 4 samples from each of the 2 tubes. In each tube there are 2 samples from the hot leg and two from the cold leg. The radiochemical concentrations (µCi/g of deposit) are consistently higher in the hot leg than in the cold leg.

In the GFP system, the PHT system sample continuously flows through the sample holder and is analyzed by the Ge detector. Periodically the detailed spectral data accumulated in the analyzer is transferred to the central computer located in the chemistry laboratory post-accident monitoring counting room. The computer is used for archiving and performing more detailed analysis. Spectral analysis is performed and stored every half an hour [Reference 6]. The Unit 2 spectral analyses data for the 1996 study period, was in the form of daily averages of the radionuclide peak area.

The graphs in Figure 2 show the trends of selected radionuclides in Unit 2 in 1996 as measured by the GFP. The activities are not expressed in absolute values (Ci /kg) but as net peak areas. The peak areas have been corrected for the efficiency curve. The benefit of this type of monitoring is for observing trends and not absolute values.

Of the particular interest is the F-17 peak area, which can conveniently be used as a reactor power indicator for monitoring trends of other radionuclides detected by GFP.

There is a general concern that the trends observed by GFP monitor may not represent the change in radionuclide concentration in the PHT system coolant, but rather the deposition of activity in the sample holder. While this concern is, to some extent a valid one, the GFP system can still be a very useful tool for providing information for activity transport monitoring. There was an endeavor to minimize possible deposition by making sample holders from materials with low affinity for fission and corrosion products deposition.

Even if there was some deposition, some trends of activated products concentration can still be observed. There is steady concentration or deposited activity of Co-60. There are discernible curves of change in concentration/deposit of other radionuclides (Sb-122/124, Zr-95, Nb-95) during the study period. Concentrations of Sb-124 and 122 are different but the curves have similar shape. The same is valid for the pair Nb-95 and Zr-95.

If deposited activity is what is dominantly recorded by GFP, it can to some extent represent a model of what is going on in the system during shutdown and operation as discussed here. In that light it can be observed from the graphs in Figure 2, that there must be significant dissolution during shutdown. Furthermore, if deposited activity is recorded, GFP monitoring results would indicate relatively stable buildup activity for all activation products but Sb-122/124. That would indicate that for all radionuclides but Sb-122/124 a deposition equilibrium is being achieved.

RADIONUCLIDE DISTRIBUTION CALCULATIONS

In this section described methods were used to evaluate activity balance and loading of radionuclides throughout the PHT system.

Activation Products

Neutron activation products come from activated corrosion (or erosion) products from PHT system materials, or ZIRCALOY fuel cladding. In this activity balance calculation, the average concentration of a specific radionuclide was used to establish the concentration of the target isotope responsible for generating the activation product.

Review of PHT Co-60 concentration data shows that there are no increasing or decreasing trends. Therefore, the average Co-60 concentration in PHT can be assumed to be constant. GFP monitoring supports this assumption.

The Co-60 activity lost (AL) from the PHT coolant, can be expressed by:

$$AL = (PAR + LAR + DPR + DAR)\, t \quad \ldots\ldots\ldots\ldots\ldots\ldots(2)$$

where:

PAR = loss rate of Co-60 through purification (Ci/h),

LAR = leak rate losses of Co-60 (Ci/h),

DPR = deposition rate of Co-60 (Ci/h),

DAR = loss rate of Co-60 through decay (Ci/h),

t = study period

Radionuclide deposition rate and purification rates are significant information for the Waste Management purposes. It can provide a tool for predicting radionuclide quantities in the deposits correlated to operational data. Purification rate provides data about radionuclide removal from the system. Removed radionuclides are distributed between filters and IX resins in the purification system. Based on chemical speciation data it can be concluded how the specific radionuclide would be distributed. In Co-60 case it is most likely a suspended particle and therefore deposited on filters.

The load of Co-60 (LCo60) on filter cartridge(s), after removal from service, can be calculated:

$$LCo60 = PAR * FE * t(F) - DAR(F) \quad \ldots\ldots\ldots(3)$$

Where:

PAR= purification loss rate (Ci/h)
FE = filter efficiency (%/100),
t(F) = time the cartridge was in service
DAR(F) = loss of activity on filter cartridge due to decay in t(F).

For the Bruce and Pickering units, all the required information for activity balance calculation was available but not for Darlington. There were no Oxiprobe results available for DNGS Unit 2. At Bruce and Pickering stations the minimum deposition rate was calculated based on the Oxiprobe data.

The example of Bruce A Unit-3 is analyzed. From Table 4 the estimate of the deposited Co-60 activity in all steam generators can be found (72 Ci). This inventory of Co-60 was estimated some time before the study but it was assumed that this was the activity at the beginning of the study period. In the study period of 126 days (90 EFPD), the Co-60 radiation fields and therefore the activity would be expected to increase to 72.6 Ci (equation 1 in section 2.3). During the same period the Co-60 would decay to 6.9 Ci, which means the loss of 3.2 Ci. As there is a net increase in steam generator fields, Co-60 deposition had to makeup 3.7 Ci in the study period of 90 EFPDs (3.2 Ci decay loss + 0.2 Ci activity increase). This information can be used to estimate the deposition rate of Co-60 in steam generators. Calculated deposition rate is $3.1 *10^{-1}$ µCi/s.

This deposition rate can be added to other loss rates from PHT coolant to obtain AL/t as described in equation (2). This loss rate is equivalent to the generation rate of Co-60 at Bruce A Unit 3. Using the equation (2) the calculated generation rate of Co-60 is $3.6 *10^{-1}$ µCi/s. Most of the Co-60 production is lost to deposition in boilers and elsewhere. The second largest loss of Co-60 produced is to the purification system.

The PNGS, Unit 1 generation rate in the period of 311 days was calculated the same way. Calculated deposition rate was 0.28 µCi/s, while the total Co-60 generation rate in Unit 1 was 0.29 µCi/s.

The out of core inventory of activation product radionuclides (applicable also for deposited fission product radionuclides) is expected to grow with operation time in accordance with the following equation [Reference 7]:

$$I = \frac{R}{\lambda}(1-e^{-\lambda\theta}) \quad ....(4)$$

where:
I= out of core radionuclide inventory (Ci)
R= production rate (Ci/s)
λ= decay constant (1/s)
θ= operational time since the circuit was new (or decontaminated) (s).

The relation in equation (4) is in general agreement with steam generator survey results that show Co-60 radiation fields increase with Unit operating time.

Equation (4) can be used to estimate the out of core inventory of Co-60. The Co-60 generation rate (R) can be calculated as AL/t from the equation (2) in this section.

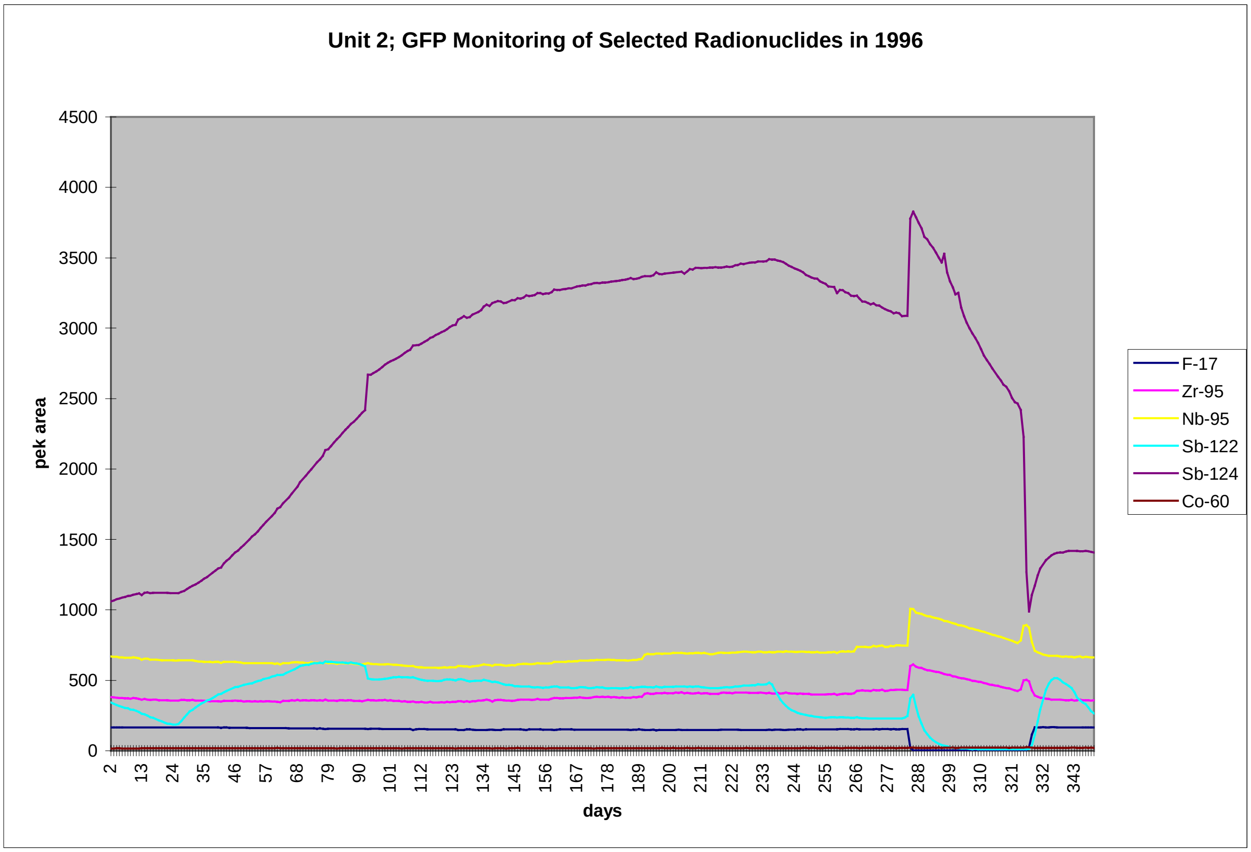

Fig. 2. Darlington Gaseous Fission Product Monitor Data

Fission Products

Fission products include those species produced directly from fission or daughters from decay in a chain of fission products. Only so called shielded radionuclides are direct fission products. The majority of fission products remain contained in the discharged spent fuel bundles, but a small portion is detected in the PHT system as a result of either diffusion or through the occasional fuel bundle defect. The produced radioactivity of a fission radionuclide (with short-lived precursors) can be calculated using Equation (5), (Reference 8).

$$A = N\lambda = F*Y(1 - e^{-\lambda Tr})\, e^{-\lambda Tc} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (5)$$

where A = activity in dps,
N = number of atoms of radionuclide (atoms),
λ = decay constant of the radionuclide (s^{-1}),
F = fission rate (fission/s),
Y = fission yield,

Tr = irradiation time (s),
Tc = time after rector shutdown- cooldown (s).

The data required for calculating the fission rate includes:

The energy generated by one thermal fission is 200 MeV.

The fission power at 92% is 2439 MW.

As discussed earlier (Section 2.1) only relatively long-lived radionuclides are considered in the PHT system activity balance. Long-lived radionuclides are those with half-lives long in comparison to reactor irradiation periods (periods between outages). Examples of relatively long lived fission products include H-3, Kr-85, Sr-90, I-129 and Cs-137. These radionuclides do not reach saturation in the reactor. There are some fission products with half-lives too long for saturation but too short for the assumption of linear build-up. Examples of these radionuclides are Ru-106, Ce-144, Pm-147.

Radionuclide Cs-137 was selected as a typical representative fission product for this demonstration activity balance because of its significance in radioactive waste management. In the following subsection, the Cs-137 radioactivity concentration based on the Cs-137 detected by radiochemistry analysis in the PHT samples is compared with the fission-generated radioactivity. The distribution of Cs-137 between spent fuel and PHT system is then evaluated.

There are three pathways for Cs-137 into the PHT system:

Diffusion through the fuel cladding,
Fuel defects,
Fission of the uranium contamination of the fuel bundle cladding. Currently, the maximum allowable U contamination in the Zircalloy is 3.5 μg/g.

It was assumed that the concentration of Cs-137, detected in the PHT system was constant during the study period. The amount of activity released into the PHT system during the study period should then be equal to the activity removed during the study period. The activity removed from the PHT system is equal to the sum of losses due to purification (PA), leaks (LA) and decay (DA). Deposition of a significant amount of Cs-137 in the system is unlikely, because chemical speciation data indicate that it is in soluble form. The radiation field surveys conducted throughout OHN stations support this statement.

The amount of Cs-137 radioactivity removed by purification (PA) during the study period is:

PA = purification rate x measured concentration in D_2O x purification period,

The Cs-137 activity removed by leakage (LA) during the study period:

LA = measured concentration in the PHT system x lost volume of D_2O in the period, The activity of Cs-137 lost through decay (DA) during the study period is:

DA = measured concentration x $(1\text{-}e^{time/T1/2})$ x mass of D_2O in PHT

The decayed activity (DA) and leak loss (LA) are negligible with respect to the amount removed by purification. The Cs-137 that is removed from the PHT system by heavy water loss (LA) is also removed in the upgrader system and IX load on upgrader IX resin can be calculated by activity balance on this system.

For the radioactive waste management perspective losses through purification where Cs-137 ends up on IX resins are the most relevant. The approximate Cs –137 load on an IX resin batch can be calculated the same way as for Co-60:

LCs137 = PA * RER* t(R) - DA(R) ……….(6)

Where

PA = purification rate x measured concentration in D_2O x purification period

RER= removal efficiency from the resin calculated from measured decontamination factor ,

t(R)= time the resin batch was in operation,

DA(R)= Cs-137 decay on the resin.

The Cs-137 generation in the observed periods at all stations was also calculated. The total amount of Cs-137 released into PHT system was typically 0.01 % compared to radioactivity Cs-137 that was removed in used fuel bundles. This indicates that there were minimal fuel defects in the systems. The amount of Cs-137 generated in one year and contained in the fuel is typically 1.5 –3 10^6 Ci in a year period. The average activity of Cs-137 in a used fuel bundle is between 510 Ci (Bruce) and 580 Ci (Pickering).

CONCLUSIONS

In this demonstration study, various sources of activity balance and transport data were reviewed. Both an activation and a fission product were used in PHT system activity balances. As a result of conducting this study, following recommendations were developed to improve data collection and usefulness of radiochemistry analysis results.

Long term sampling programs have to be modified and adjusted to obtain better information for radioactivity transport analysis.

Sampling should be performed at locations where there is expected to be a change in radionuclide concentration. Three locations are recommended in CANDU stations:

PHT Main Circuit samples to represent reactor outlet samples.

Common inlet of bleed filters to represent reactor inlet and purification inlet samples.
Outlet of operating ion exchange column to represent purification outlet samples.

The samples should be taken as close in time as possible. All samples should be taken weekly until it can be demonstrated that a less frequent sampling schedule is satisfactory. This will occur when the relation of results between sampling points is established. Results should be compared and ratios established. Purification system removal efficiency should be calculated.

Large 1 L samples should also occasionally be taken. Samples should be treated with Millipore and ion exchange filters and analyzed to obtain chemical speciation information.

A frequency of 3-4 CRUD samples a year is required. Results should be compared to results of radiochemistry of large 1 L taken during transient operation.

Radiation field survey conducted on equipment where CRUD is expected to deposit would be very beneficial however, total gamma fields must be monitored. CRUD typically deposits in locations where there is a low temperature in the PHT system.

Activity balance and transport data must be reviewed once a year or as a minimum once every two years.

GFP monitor would be more useful if it was calibrated to activities (μCi/kg). This would make data more useful for activity balance purposes.

Oxiprobe measurements and results is one of the most valuable tool in assessing deposits in steam generators. The most thorough radiochemistry analysis to determine the amount of α and β emitters can be performed. Radioactivity deposited in steam generators is removed by decontamination and it ends up on filters or IX resins. The transfer of α and β emitters to cleaning media in such activities could be assessed.

REFERENCES

1. Bruce A: PHT System Activity Transport Monitoring Report. Report No.:NK2101811 CMD 965142 R00, January 1997.

2. PNGS A: PHT System Activity Transport Monitoring Report. Draft Report No.:NA44 01811P CMD 965 169R00, December 1996.

3. DNGS: PHT System Activity Transport Monitoring Report. Draft Report No. NK38-33200-CMD-P, November 18, 1997.

4. F. Gonzalez et al.: Oxiprobe Measurements at Bruce NGS Unit 2, OH Report: A-NSG-95-163-P .

5. F. Gonzalez et al.: Oxiprobe Measurements at PNGS U-1, SG-8 Primary Side Tube Deposit and Radionuclide Activity Determinations. OH Report No.A-NSG-95-162-P, March 1996.

6. Z. Lovasic, personal communication with S. Lim.

7. Activity Transport Manual, AECL, WNRE-389.

8. M. Benedict, T. Pigford and H. Levi: ”Nuclear Chemical Engineering”, McGraw-Hill 1981.

9. K.A Burril: Deposition of Corrosion Products in Core, 2nd CANDU Chemists Workshop, Sept 1996.