GAMMA SPECTROMETRY MEASUREMENTS FOR MONITORING ACTIVITY TRANSPORT AT THE NUCLEAR STATIONS

by

Aamir Husain
Environmental and Nuclear Services
Kinectrics

ABSTRACT

The deposition of activation and fission products on the out-core surfaces of the CANDU primary heat transport and moderator systems leads to growth of gamma radiation fields around piping and other system components. It is desirable, for the purposes of monitoring radiation field growth, to routinely conduct in-situ gamma spectrometry of the various system components such as the reactor face, feeder cabinets, boilers and moderator heat exchangers. This paper describes various applications involving the use of both hyperpure germanium and cadmium zinc telluride detector systems. The methodology used to interpret the measured data is also outlined.

1.0 INTRODUCTION AND BACKGROUND

The deposition of activation and fission products on the out-core surfaces of a CANDU reactor leads to the growth of gamma radiation fields around various primary heat transport (PHT) and moderator system components. The deposits are responsible for the radiation doses received by workers undertaking inspection and maintenance tasks during reactor shutdown.

Activation products in the PHT system arise from the corrosion of system materials, which release oxidation products to the circulating coolant either as dissolved metal ions or as particulate/colloidal metal oxides. Transport and deposition of these products in-core, their subsequent activation in the neutron flux and reentry of the activated products into the coolant results in contamination of the out-core surfaces. Among the principal activation products observed on PHT system surfaces, namely, Co-60, Zr-95, Nb-95, Cr-51, Fe-59, Mn-54 and Zn-65, the dominant contributor to out-core radiation fields is generally due to Co-60. On the other hand, fission products on out-core surfaces arise from the transport of radionuclides which escape from defects in the fuel element cladding. The gamma emitting fission products generally observed on out-core PHT system surfaces include Zr-95, Nb-95, Ba-140 and La-140.

As in the case of the PHT system, fields around the moderator system also arise from deposited activation and fission products. Activation products such as Tb-160 and Sc-46 arise from the activation of contaminants introduced along with consumables into the moderator system while radionuclides such as Co-60, Cr-51 and Zr-95/Nb-95 result from the corrosion of system materials (Zircaloy-2, stainless steel and satellites) followed by subsequent activation of the corrosion products. Fission products are also observed in the moderator system as a result of the trace levels of uranium present in Zircaloy-2.

Table 1 presents some particulars for selected activation and fission products observed in the PHT and moderator systems.

Table 1 Principal Activation and Fission Products Observed in Gamma Spectra of CANDU Reactor System Components

Radionuclide	Halflife	Production Reaction	Principle Gamma Energy (MeV)
Sc-46	84 d	Sc-45(n-γ)	1.121, 0.889
Cr-51	28 d	Cr-50(n-p)	0.320
Mn-54	310 d	Fe-54(n-p)	0.835
Co-58	72 d	Ni-58(n-p)	0.811
Fe-59	45 d	Fe-58(n-γ)	1.292, 1.099
Co-60	5.3 y	Co-59(n-γ)	1.332, 1.172
Zn-65	244 d	Zn-64(n-γ)	1.115
Zr-95	65 d	Zr-94(n-γ) & Fission	0.756, 0.724
Nb-95	35 d	Zr-95 Decay & Fission	0.766
Ru-103	39.5 d	Fission	0.497
Ru-106	1 y	Fission	0.622
Sb-124	60 d	Sb-123(n-γ)	1.692
Ba-140	12.8 d	Fission	0.537
La-140	40.3 h	Fission	1.596
Gd-153	241.6 d	Gd-152(n-γ)	0.0975, 0.103
Tb-160	73 d	Tb-159(n-γ)	0.876, 0.299

It is desirable, for the purposes of monitoring radiation field growth, to routinely conduct in-situ gamma spectrometry of PHT and moderator system components. Such surveys permit the contributions of various radionuclides to the radiation fields to be trended with reactor operation. This is valuable for monitoring the effects of chemistry changes on activity transport within the PHT and Moderator systems and also in decontamination planning.

In-situ gamma spectrometry of reactor system components at Pickering and Bruce nuclear stations were routinely conducted until about the mid 1990s. They were then discontinued for a period of several years before being re-started at Bruce Power in 2003. Several units at both Bruce Stations A and B have been surveyed since then. Ontario Power Generation has also shown renewed interests in outage surveys with the first set of Darlington scans being recently completed at Unit 3. Typically, the components of interest include the reactor face, feeder cabinets, boilers and moderator heat exchangers.

Gamma spectroscopy is typically performed using a shielded/collimated hyperpure germanium detector system. Germanium detectors offer a high peak resolution and are capable of adequately resolving the peaks associated with most observed radionuclides. In some applications, a lower degree of resolution is acceptable and use of cadmium zinc telluride detectors (CdZnTe) may be considered. These detectors, in contrast to the germanium detectors, do not require to be liquid nitrogen cooled and can operate at room temperature. Because of their smaller size, they can be utilized in much higher radiation field environments (up to several rem/h).

This paper presents an overview of the application of germanium detector systems for the monitoring of activity transport in CANDU reactor systems. The methods used for interpreting gamma spectra are discussed and typical results are presented. Specialised applications including those based on CdZnTe detectors are also presented.

2.0 INTERPRETATION OF GAMMA SPECTROMETRY SURVEY RESULTS

Typically, the objective of performing activity monitoring surveys is to apportion the measured radiation field from a component into its radionuclide specific contributions. Thus, by trending, say the Co-60 contribution to the reactor face field, one can attempt to understand the reasons for its decline or increase between two successive outages. Interpretation of the gamma spectra and associated radiation field results in this manner does not require an estimation of radionuclide activities present within the component but rather the effect of this activity on the external radiation fields. In some cases, however, determination of the activity within a component may be of interest. Both interpretations require that the component of interest be represented by a simplified geometry that is amenable to modeling.

Although the radionuclides present in a spectrum can be readily identified and count rates determined (see Figure 1), the quantitative interpretation of the results in terms of radiation field compositions or radionuclide activities within a component is not straightforward. This is because the incident gamma ray flux is attenuated by the various shielding materials present and also undergoes Compton[1] scattering resulting in the production of secondary photons. The latter also contribute to the external gamma field, a phenomenon which is termed 'buildup'. The magnitudes of the shielding and buildup effects are energy dependent. The consequence of buildup can be observed in field spectra where, in contrast to typical laboratory spectra for unshielded sources, the background can be observed to increase dramatically with decrease in photon energy.

Gamma spectra for boilers at OPG and BP plants have historically been interpreted according to the methodology developed by Spekkens [1979]. The methodology estimates the contribution of each radionuclide to the external boiler radiation fields by considering the tube bundle to be a uniformly distributed cylindrical source, which is shielded from the external detector by several intervening layers of metal, water and insulation. A dose build-up factor is incorporated to account for Compton scatter in the shielding layers. The formulation accounts for all the emissions associated with the decay of each nuclide, including those, which are not observed in the gamma spectra. Accordingly, the radiation field contribution F_A of a nuclide A is given by the expression

$$F_A = K \frac{N_1}{\varepsilon_1}\left(B_1 E_1 + B_2 \frac{A_2}{A_1}\frac{I_2}{I_1} E_2 + B_3 \frac{A_3}{A_1}\frac{I_3}{I_1} E_3 + ... \right) \qquad (1)$$

where

K is a constant

[1] Compton scatter refers to a specific type of interaction of the incident gamma ray with the electrons associated with the atoms of the shielding material, whereby the electron is ejected from the atom and the incident gamma ray is scattered with a lower energy.

N_1 is the count rate at peak energy E_1
ε_1 is the efficiency of the detector at peak energy E_1
B is the dose buildup factor, and
A is the photon abundance, and
I is the geometry factor.

Subscripts 1, 2, 3…refer to the multiple emission energies of nuclide A. The percent field contribution of nuclide A is then obtained from the ratio $F_A / (F_A + F_B + F_C + ...)$.

Figure 1: Typical Reactor Face Spectrum Obtained Using a Germanium Detector System

The parameters in Equation (1) can be readily calculated based on the dimensions of a boiler. Values for Bruce and Pickering boilers have thus been estimated. While the equation was derived for a cylindrical boiler source, it appears intuitively to be valid for a general component provided the appropriate values for buildup and geometry factors can be estimated. Because of the difficulty in estimating these factors generally, Equation (1) has been historically used to interpret reactor face and feeder cabinet spectra assuming the ratios B_2/B_1, B_3/B_1,.. I_2/I_1, I_3/I_1 …to be all equal to unity. This simply amounts to the neglect of buildup and shielding effects while still fully accounting for all peak emissions from each individual radionuclide.

Equation (1) is based on an analytical expression for the external dose rate. With the availability of shielding codes such as MicroShield, a somewhat different approach can be more generally applied. The latter also assumes that components such as boilers or heat exchangers can be modeled as a uniformly distributed cylindrical source surrounded by several layers of shielding materials. This approach is now presented for the case of a Bruce B moderator heat exchanger [Husain et al 2005].

2.1 Interpretation of Moderator Heat Exchanger Spectra

The moderator heat exchangers at Bruce have a horizontal shell and tube configuration. The incoming heavy water flows through the primary side tubing, making four passes across the length of the heat exchanger, before exiting. It is cooled by light water on the secondary side.

The total radiation field *F* at a selected distance from the surface of the moderator heat exchanger can be generally expressed as follows:

$$F = F_A + F_B + F_C + \ldots \quad (2)$$

where F_A, the radiation field arising from radionuclide *A* can be expressed as

$$F_A = f_A C_A \quad (3)$$

f_A is the radiation field per unit activity for nuclide *A* and C_A its activity in the source. Further, C_A can be expressed as

$$C_A = N_{A_{E1}} / (\varepsilon_{A_{E1}} A_{A_{E1}}) \quad (4)$$

where $N_{A_{E1}}$, $\varepsilon_{A_{E1}}$ and $A_{A_{E1}}$, respectively, represent the count rate, detector efficiency and abundance for a peak with emission energy *E1* arising from the gamma decay of radionuclide *A*. For known values of $f_A, f_B, f_C\ldots$, the above expressions can be combined to determine the percent contribution of individual radionuclides. In practice, a point source efficiency is used instead of the actual source efficiency and hence a proportionality constant must be substituted for the equality sign in Equation (4).

Values of the radiation field per unit activity for the individual radionuclides present in the source, i.e., $f_A, f_B, f_C\ldots$ can be determined using the MicroShield code considering only the straight section of the tube bundle. This requires a determination of the attenuation coefficient of the source material μ_s. The latter is obtained by considering the unit cell in Figure 1 which represents the layout of the tubes in the tubesheet based on a triangular pitch. The unit cell, a repeat unit across the tube bundle, thus effectively consists of metal corresponding to the cross-section of half a tube with the balance consisting of primary and secondary side water. Accordingly, the attenuation coefficient of the source tube bundle material μ_s was estimated from the expression $\mu_s = 1.4\ (\mu/\rho)_{metal} + 0.841(\mu/\rho)_{water}$ where ρ denotes density.

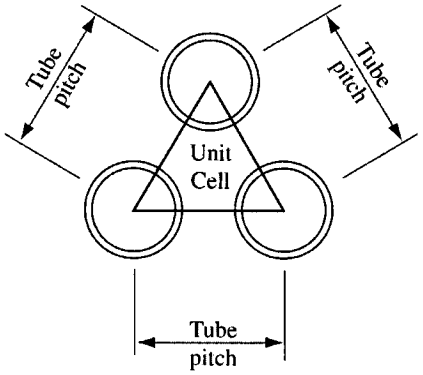

Figure 2: Tube Layout Showing Unit Cell

MicroShield calculations were next performed using μ_s and considering, in accordance with the design of the heat exchanger, a cylindrical source of approximate height 198 inch and radius 34 inch surrounded by a secondary side water layer of approximately1.93 inch and a steel shell of thickness 0.875 inch. The phenomenon of dose buildup was taken into account in estimating radiation fields. The radiation field due to 1 Bq activity for each radionuclide of interest, i.e., f_A, f_B, f_C etc, were calculated at various external dose points. In general, their relative values were found to be insensitive to the location of the dose point.

Although the calculations suggest that the interpretation of the gamma scan results would be insensitive to the specific location of the gamma detector, the conclusion is based on a simplified heat exchanger geometry which neglects the internal structures present on the secondary side. Considering the latter, it is preferable to locate the detector at mid length of the heat exchanger, with its axis perpendicular to the horizontal axis of the heat exchanger, thus minimising the shielding effect of the baffle plates which are spaced at regular intervals across its length.

Interpretation of a Bruce Unit 5 moderator heat exchanger spectrum, collected as described above, is illustrated in Table 2. Because of the use of point source efficiency data, values for C_A and F_A as shown are correct to within a factor K where K can be determined by comparing the ΣF_A with the measured field at the location of the detector. Note that the % Contribution given in the last column of Table 2 is independent of the factor K.

2.2 Other General Considerations

Decay Correction

Typically, the field contributions are decay corrected to 10 days after shutdown in order that results from various outages can be consistently compared. Regardless of this, it is desirable to obtain the field spectra soon after shutdown when some of the shorter lived radionuclides such as Cr-51 are still detectable.

A preferred window for performing the measurements is within 10 days after shutdown. For comparison of results between outages, particularly when the spectra are not

consistently acquired within a short time window after shutdown, it is strictly valid to compare only the actual magnitudes of the radiation field which are attributed to individual radionuclides. Direct comparison of the % field contribution data between outages is risky in such cases because the values may be distorted by the presence of short-lived radionuclides in some measurements and their absence in other measurements. Unfortunately, one may be constrained by other factors such as the appearance of hot spots on the reactor face to be able to only compare the % field contributions between outages.

Table 2: Contribution of Various Radionuclides to Bruce Unit 5 Moderator Heat Exchanger Fields - Calculations Illustrating the Application of Equations (1)-(3)

Nuclide	Peak Energy E1 (keV)	Detector Efficiency $\varepsilon_{A_{E1}}$	% Abundance $A_{A_{E1}}$	Peak Area* $N_{A_{E1}}$	Radiation Field per Unit Activity f_A	Activity C_A	Radiation Field F_A	% Radiation Field Contribution
Co-60	1332	1.62E-05	100.0	3390	7.17E-11	2.1E+06	1.50E-04	6.1E+01
Fe-59	1292	1.69E-05	43.2	324	3.31E-11	4.4E+05	1.47E-05	6.0E+00
Nb-95	766	3.70E-05	100.0	5299	1.76E-11	1.4E+06	2.51E-05	1.0E+01
Zr-95	756	3.78E-05	54.8	1827	1.66E-11	8.8E+05	1.47E-05	6.0E+00
Cr-51	320	1.37E-04	9.8	3965	4.04E-13	3.0E+06	1.19E-06	4.9E-01
Tb-160	876	3.03E-05	30.0	1099	2.62E-11	1.2E+06	3.17E-05	1.3E+01
Sc-46	1121	2.09E-05	100.0	286	5.26E-11	1.4E+05	7.19E-06	2.9E+00

*Decay corrected 10 prior to shutdown

Effect of Boiler Draining

Boilers may be in a drained state during measurements with one or both sides (primary and secondary) of it being drained. Draining results in reduced shielding and hence an increase in measured radiation fields. Historically, boiler dose rate measurements have been corrected using a Drained Boiler Factor to convert the measurements into Equivalent Full Boiler (EFB) fields. This facilitates comparison of the data between outages. For Bruce boilers, the DBF values corresponding to boilers with only primary side drained and boilers with both primary and secondary sides drained are 1.1 and 3.1, respectively. These factors are borne out by measured data and are also easily calculated using MicroShield.

Similar to the effect on boiler dose rates, draining, particularly of the secondary side, would exaggerate the peak count rates of radionuclides with lower emission energies. Historically, this effect has been ignored when interpreting boiler spectra with the same set of shielding and buildup factors being used regardless of the drained state of the boiler. This gap should be addressed to obtain more meaningful data and hence to improve data trending.

3.0 RESULTS FROM SELECTED GAMMA SPECTROMETRY APPLICATIONS

3.1 Recent Surveys at Bruce Unit 5

Gamma spectrometry and dose rate surveys were recently (November 2005) performed at Bruce Unit 5. Of special interest were the boilers because half of them had been

mechanically cleaned just prior to the measurements. Boiler survey results, based on contact radiation fields measured at up to 8 locations around each boiler (Elevation 678 feet), are summarised in Table 3. Because the primary and secondary sides of all the boilers were drained at the time of measurement, a drained boiler correction factor of 3.1 was applied to convert the data into EFB fields. No correction for decay between the measurement date and the date corresponding to 10 days after shutdown was necessary because gamma spectrometry indicated the fields were essentially due to the relatively long-lived Co-60.

As shown in Table 3, the EFB radiation fields for the mechanically cleaned boilers, namely, Boilers 2, 3, 6, and 7 ranged between 0.9 and 1.4 mrem/h whereas the fields for Boilers 1, 4, 5 and 8 which were not cleaned ranged between 5.1-6.0 mrem/h. This indicates a decontamination factor of 4.9 or equivalently an oxide removal efficiency of approximately 80% was achieved. Independent assessment of the oxide removal efficiency based on initial estimates of oxide loading in the boilers and the amount of magnetite waste produced during the clean [Wilson 2006] are consistent with this estimate.

As in the case of the boilers, Co-60 was also found to be the dominant contributor to the fields in the reactor vault. Based on reactor face and feeder cabinet measurements, its contribution was found to vary between 83 and 97% with the balance being due to Zr-95 (2.3-5.8), Nb-95 (3.3-9.3 %) and Sb-124 (0-3 %). Because reactor vault spectra are interpreted without considering shielding and buildup effects, the contributions for radionuclides with lower emission energies such as Zr-95 and Nb-95 are under-estimated relative to the contributions for Co-60. Generally, the measured ratio of Zr-95/Nb-95 was consistent with the ratio of 0.46 expected for this parent daughter pair at equilibrium. Comparison of the spectral data for the reactor vault and boilers indicates that the relatively insoluble Zr-95 and Nb-95 largely deposit out on system surfaces before they have a chance to make it to the boilers.

Table 3: Summary of Measured Boiler Radiation Fields

Boiler	Field As Measured (mrem/h)	Equivalent Full Boiler Radiation Field (mrem/h)
1	16.1 ± 2.0	5.2 ± 0.6
2	3.5 ± 0.9	1.1 ± 0.3
3	3.3 ± 1.0	0.9 ± 0.5
4	18.4 ± 1.9	5.9 ± 0.6
5	15.6 ± 1.5	5.0 ± 0.5
6	2.8 ± 0.9	0.9 ± 0.3
7	4.2 ± 0.9	1.4 ± 0.3
8	17.3 ± 0.9	5.6 ± 0.8

Compared with the field compositions in the vault and at the boilers, moderator heat exchanger fields were also dominated by Co-60, although its contribution was only 43%.

Significant levels of Zr-95 (6.9%), Nb-95 (13.7%), Tb-160 (17.0%) and Sc-46 (7.4%) were observed.

3.2 Applications During Primary Heat Transport System Decontaminations

Several chemical decontaminations of the PHT systems at Pickering Units 1-4 were conducted during the period 1980 to 1994 to primarily reduce the radiation fields at the reactor face in preparation for retubing. While the earlier decontaminations employed only a reducing (R) CAN-DECON solvent, the later decontaminations also employed an oxidising (O) dilute alkaline permanganate step to condition the chromium-rich oxides on stainless steel end-fittings.

Typically, gamma spectrometry of various PHT system components was performed in support of the decontaminations and complemented the extensive dose rate measurements. The components surveyed included the reactor face, feeder cabinets, boilers and end shield cooling piping. Scans at these locations were collected every half hour in order to monitor the progress of the decontaminations. Typically, while the Co-60 count rates at the reactor face trended downwards, Sb-124 count rates were observed to increase as a result of its transport out of the boilers and the reactor core.

In addition to gamma spectrometry at the afore-mentioned locations, selected feeder pipes were also monitored during the 3-step (ROR) 1989 Pickering Unit 3 and the 1991 Unit 4 one-step (R) decontaminations. Measurements were performed inside the horizontal feeder cabinets where the radiation fields ranged up to 525 mrem/h. The spectrometer was located across the walkway during the measurements with the horizontal axis through the detector intersecting the vertical axis of the chosen pipe. In order to selectively monitor the pipe of interest, the detector was suitably collimated to eliminate the contributions of adjacent feeder banks and a shield was employed behind the pipe to minimise the contributions from the pipes in the same bank which were behind it. Measurements were taken with and without a solid lead plug inserted into the opening of the collimator; this allowed the significant background counts to be accounted for in the data interpretation. The net count rates obtained thus were converted into equivalent radionuclide activities based on a validated pipe source efficiency calibration methodology; the latter considered a pipe to be equivalent to a system of two parallel plates separated by its internal diameter.

Results for an inlet and an outlet feeder pipe obtained during the 1989 Pickering Unit 3 decontamination [Husain 1995] are shown in Table 4. Co-60 as expected was the principal source of activity prior to the decontamination. As a result of the oxide dissolution in the R step, Co-60 activity, particularly on the inlet feeder was significantly reduced while out-of-core transport caused the Sb-124 activity on the outlet feeders to increase markedly. Increase in Sb-124 activity on the inlet feeder after the R step was possibly caused by transport from the boilers. Based on the results for the P1 outlet feeder, additional activity transport occurred in the intervening 45 days between the first R step and the O step possibly due to defueling/draining activities.

3.3 Application of CdZnTe Detectors in Boilers

During normal outage surveys, acquisition of boiler spectra using the portable germanium detector is feasible only at one specific elevation because of access considerations. Further, only one side of the boiler is generally accessible at this

elevation. Because of the need to shield the detector, the spectra obtained reflect the activity from a small slice of the boiler with activity from both the cold and the hot leg being measured.

Table 4: Measured Activity of Feeder Pipes During Pickering Unit 3 Decontamination

Nuclide	Activity of Outlet Feeder P1 (mCi/m^2)				Activity of Inlet Feeder T18 (mCi/m^2)			
	Before R	After R	Before O	After ROR	Before R	After R	Before O	After ROR
Ru-103	-	1.2	-	-	2.5	-	-	-
Zr-95	1.8	-	-	-	-	-	-	-
Nb-95	2.7	-	-	-	4.9	-	-	-
Fe-59	0.8	-	-	-	1.9	-	-	-
Co-60	5.4	3.0	4.8	1.1	56.6	1.5	-	0.84
La-140	0.54	-	-	-	0.72	-	-	-
Sb-124	0.27	2.8	1.0	0.72	0.48	1.7	-	1.2
Field (mrem/h)	72	50	55	19	525	37	40	19

In contrast to the above, the use of miniature CdZnTe detectors can provide a distribution of activity as a function of tube length. For this purpose, the detector must be inserted through the tubesheet into a central tube either in the cold or the hot leg. Because the detector's response is derived primarily from a horizontal slice of several hundred surrounding tubes of approximately 50 cm length, repeating the measurements at tube locations greater than 50 cm apart provides the activity response as a function of tube length.

Measurements as described above were taken at several Pickering and Darlington boilers in support of the development of SGSCAN, a technique for measuring magnetite loading within boilers. The results obtained also provide an interesting insight into the distribution of activity within boiler tubes. Typically, Co-60, Zr-95 and Nb-95 were the only radionuclides observed (see Figure 3).

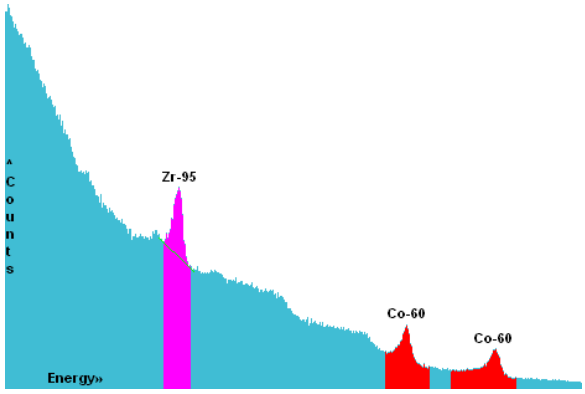

Figure 3: Response of CdZnTe Detector within a Boiler

Results for one Pickering and one Darlington campaign are shown in Figure 4. These indicate the following:

- Co-60 activity in the Pickering boiler manifests a distinctly different behavior with increasing tube length (measured from the hot leg primary side tubesheet face), compared with that in the Darlington boiler. In both cases, however, the Co-60 activity experienced an increase with tube length in the preheater region (>1500 cm) with the increase at Pickering being more dramatic. Variation of Co-60 activity was generally found to mimic the variation of oxide loading with tube length.

- Although both Zr-95 and Nb-95 were observed, they were not necessarily observed together as would be expected for a parent-daughter pair. Zr-95 appeared to decrease sharply in the preheater region of the Pickering boiler while Nb-95 manifested the reverse trend in the Darlington boiler.

4.0 CONCLUSIONS

Kinectrics has been involved in various gamma spectrometry applications at OPG and Bruce Power stations for several years. The applications have included routine surveys conducted during outages for the purpose of trending component fields, applications during PHT system decontaminations for monitoring their progress and specialized spectrometry based on the novel, miniature CdZnTe detectors.

With plans for increased feeder pipe replacement at OPG and because of the significant ramping up of reactor face radiation fields at Bruce Power, there is an increased need for gamma spectrometry support. As plans are developed to address these difficult issues, consideration should be given to the use of the CdZnTe detectors which because of their small size may be particularly useful for identifying the causes for hot spots at the reactor face.

ACKNOWLEDGEMENTS

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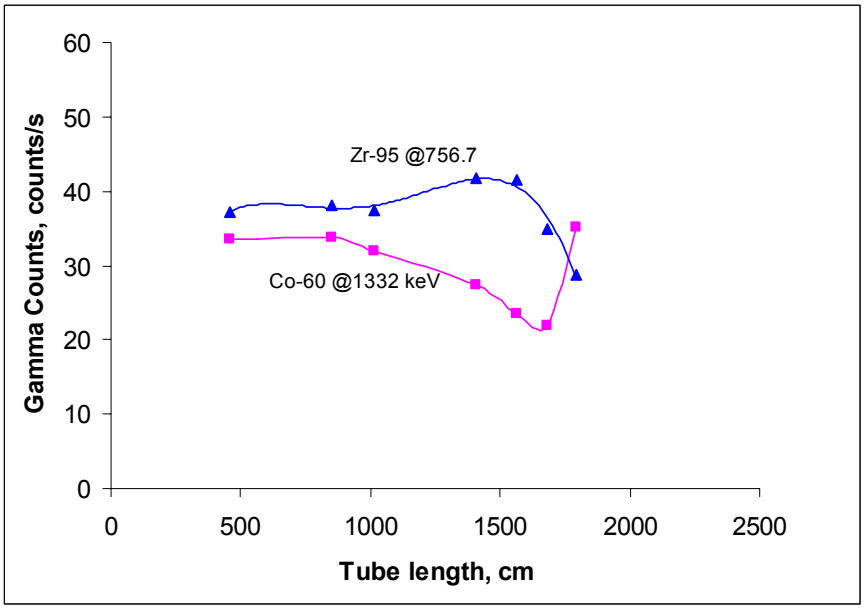
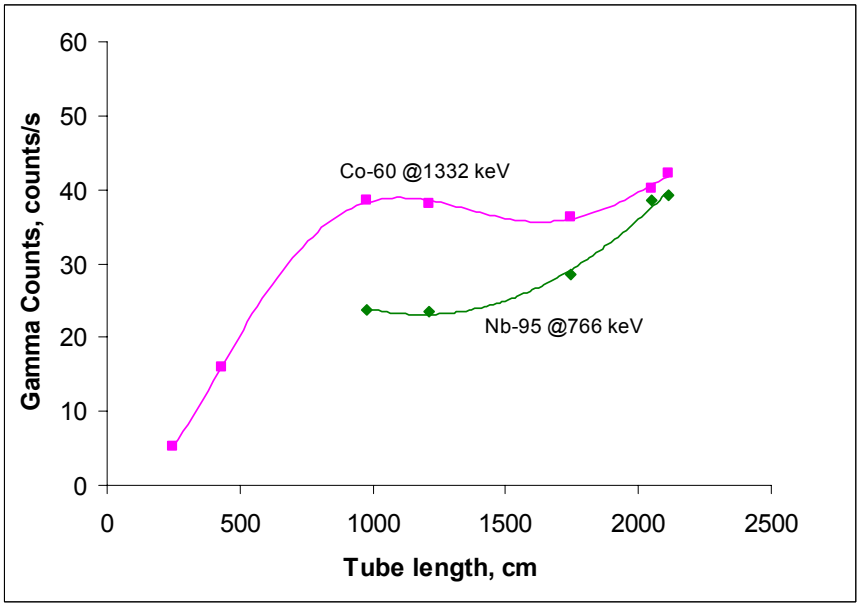


(a) Measurements taken in PNGS U8 SG 8 (Tube R34C40) in 2001



(b) Measurements taken in DNGS U 2 SG1 (Tube R46C48) in 2000

Figure 4: Results Obtained Using a CdZnTec Detector Inside a Steam Generator Tube

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