

## **CHARACTERIZATION OF ONTARIO POWER GENERATION'S LOW AND INTERMEDIATE LEVEL REACTOR WASTE – AN OVERVIEW**

A. Husain  
Nuclear Waste and Tritium Solutions  
Environmental and Nuclear Services  
Kinectrics, 800 Kipling Av., Toronto M8Z 6C4.

### **ABSTRACT**

Low and intermediate level wastes from Ontario Power Generation (OPG)'s nuclear operations are currently stored at its Western Waste Management Facility in Bruce. Eventually, these wastes will be permanently disposed in a repository. Data on radionuclide inventories in the waste are required in sufficient detail to support the development of conceptual designs, safety assessment and licensing of the planned repository. The radionuclides of interest include typical easy-to-measure gamma emitters such as Co-60 and Cs-137 and various difficult-to-measure (DTM),  $\alpha$  and  $\beta$  emitting radionuclides.

Kinectrics has been involved, for several years, in the radiochemical, chemical and microbiological characterization of OPG's reactor waste. This overview paper will focus on the radiochemical characterization of OPG's waste with emphasis on the more recent activities. The latter include 1) measurement of gamma activity in large waste packages, 2) application of dose rate-to-gamma activity conversion methodology, 3) development of scaling factors for DTM radionuclides and 4) development of predictive models for estimating scaling factors.

### **1.0 INTRODUCTION AND BACKGROUND**

Low Level Waste (LLW) produced from Ontario Power Generation (OPG)'s nuclear operations consists largely of bulk non-processible, compactible and incinerable wastes, and resin and filter waste from systems such as the Irradiated Fuel Bay purification system and the Active Liquid Waste Treatment System (ALWTS). Compactible wastes and incinerable wastes are volume reduced by box compaction and incineration, respectively. Bottom ash and baghouse ash are generated as a result of the incineration. Containers filled with various types of processed and unprocessed LLW are currently stored within the Low Level Storage Buildings at OPG's Western Waste Management Facility (WWMF).

Intermediate Level Waste (ILW) produced from OPG's nuclear operations consists principally of resin and filter wastes originating from the Moderator and Primary Heat Transport (PHT) coolant purification systems and to a lesser extent of irradiated core components. The ILW contains significantly higher activity levels than the LLW.

For the long-term management of radioactive waste, OPG plans to dispose the LLW and the ILW in a repository. Such a repository may be sited in future adjacent to the WWMF. Data on radionuclide inventories in the waste are required in sufficient detail to support the development of conceptual designs, safety assessment and licensing of the planned repository. The

radionuclides of interest include typical easy-to-measure (ETM) gamma emitters such as Co-60 and Cs-137 and various  $\alpha$  and  $\beta$  emitting, difficult-to-measure (DTM) radionuclides.

Safety assessments of OPG's repository concepts have identified several potentially critical  $\alpha$  and  $\beta$  emitting radionuclides. These include C-14, I-129, Cl-36, Nb-94, Tc-99, Se-79, Np-237, Ni-59, H-3, and isotopes of U, Th, Pu, Pa and Am. In addition, the operational safety assessment of the WWMF has identified Pu-241, a beta emitter, to be also an important radionuclide.

Kinectrics has been involved, for several years, in the radiochemical, chemical and microbiological characterization of OPG's reactor waste. This overview paper will focus on the radiochemical characterization of the waste with emphasis on the more recent activities. The latter include the following:

1. Measurement of gamma activity in large waste packages,
2. Application of dose rate-to-gamma activity conversion methodology,
3. Development of scaling factors<sup>1</sup> for DTM radionuclides,
4. Development of predictive models for estimating scaling factors, and
5. Development of analytical capability to measure DTM radionuclides.

An overview of items 1-4 is presented next. Item 5 will not be discussed here. However, it should be noted that Kinectrics currently has analytical capability for several DTM radionuclides including Pu and Cm isotopes, Am-241, Fe-55, Sr-90, C-14, Ni-63, I-129 and Cl-36. Also, method development for Tc-99 is currently underway.

## **2.0 MEASUREMENT OF GAMMA ACTIVITY OF LARGE WASTE PACKAGES**

Among the various types of wastes stored at the WWMF, waste streams such as incinerator ash (bottom and baghouse ash) and spent resins (in particular the low activity resins) are relatively easy to sample; the samples can then be characterized in the laboratory for gamma activity. On the other hand, representative sampling of non-processible and compactible wastes is difficult because of their heterogeneity. In-situ gamma counting of individual bags of these wastes can, however, be performed by placing the bags on a rotating stage during a count thus reducing effects due to heterogeneity. Both approaches, namely gamma counting of waste samples and waste bags, have been utilized but have limited applicability when large volumes of wastes are involved. In general, a capability for in-situ gamma counting of large waste packages is desirable and has wider applicability. Some applications are briefly discussed below.

Gamma activity data for 100 drums containing assorted non-processible wastes were developed in 1998 using Canberra's Q<sup>2</sup> drum scanning system. The trailer mounted system consisted of a shielded turntable, shielded germanium detectors stacked vertically to cover the height of the drum, electronics, computer and calibration drums. For counting, the drum was placed on a turntable which automatically rotated when the shielded door was closed. Strain gauges mounted on the turntable structure automatically weighed the drum. The weight of the drum contents was used to select the appropriate calibration data in order to appropriately correct for matrix attenuation. Available drum calibration standards ranged in density between 0.034 g/cc and 1.6 g/cc.

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<sup>1</sup> Scaling factors relate the activity of DTM radionuclides to the activity of easy-to-measure (ETM) gamma-emitting marker radionuclides such as Co-60 and Cs-137.

Recently, 20 waste packages were gamma scanned using Kinectrics' portable hyperpure germanium gamma scanner. The packages consisted of 2.5 m<sup>3</sup> ash bins (1.32x1.32x1.42m), 2.8 m<sup>3</sup> B25 compactor boxes (1.84 x 1.17x1.2m) and 3.0 m<sup>3</sup> non-processible waste containers (1.96x1.32x1.19m). The waste densities ranged from 0.24 g/cc to 0.68 g/cc. Because of the impracticality of calibrating large waste boxes using known box sources, specialised software called ISOTOPIC v3.1 was procured from Ametec (formerly Ortec); this software permitted calibration data for any box source to be derived from the readily obtained point source calibration curve. Figure 1 illustrates the setup during one measurement; a weigh scale (visible in the background) was used to determine the density of the waste which, in turn, was used to estimate the appropriate box source calibration data. Because of the large size of the boxes, scans were performed on two opposite sides (large face) of each box and the results averaged. The measured activities were consistent with past data for these wastes obtained by counting samples or individual bags.



**Figure 1 In-Situ Scanning of an Ash Box Using the Portable Germanium Detector**

The portable detector system was also used recently to assay two drums containing fuel bay filters from Darlington. The filters were approximately 3 ft in height and 1 ft in diameter. In contrast to the large waste boxes which had relatively low dose rates (< 5 mRem/h), the filters had fields exceeding 100 mRem/h. Photographs of the filters are shown in Figure 2. Results obtained by in-situ scanning were validated by a) verifying the activity of a known Cs-137 source placed at the middle of an empty drum, b) comparing the in-situ results with scaled up results for samples removed from the filter and c) comparing dose rates, estimated from the measured activity data, with measured dose rates.



(a)



(b)



(c)

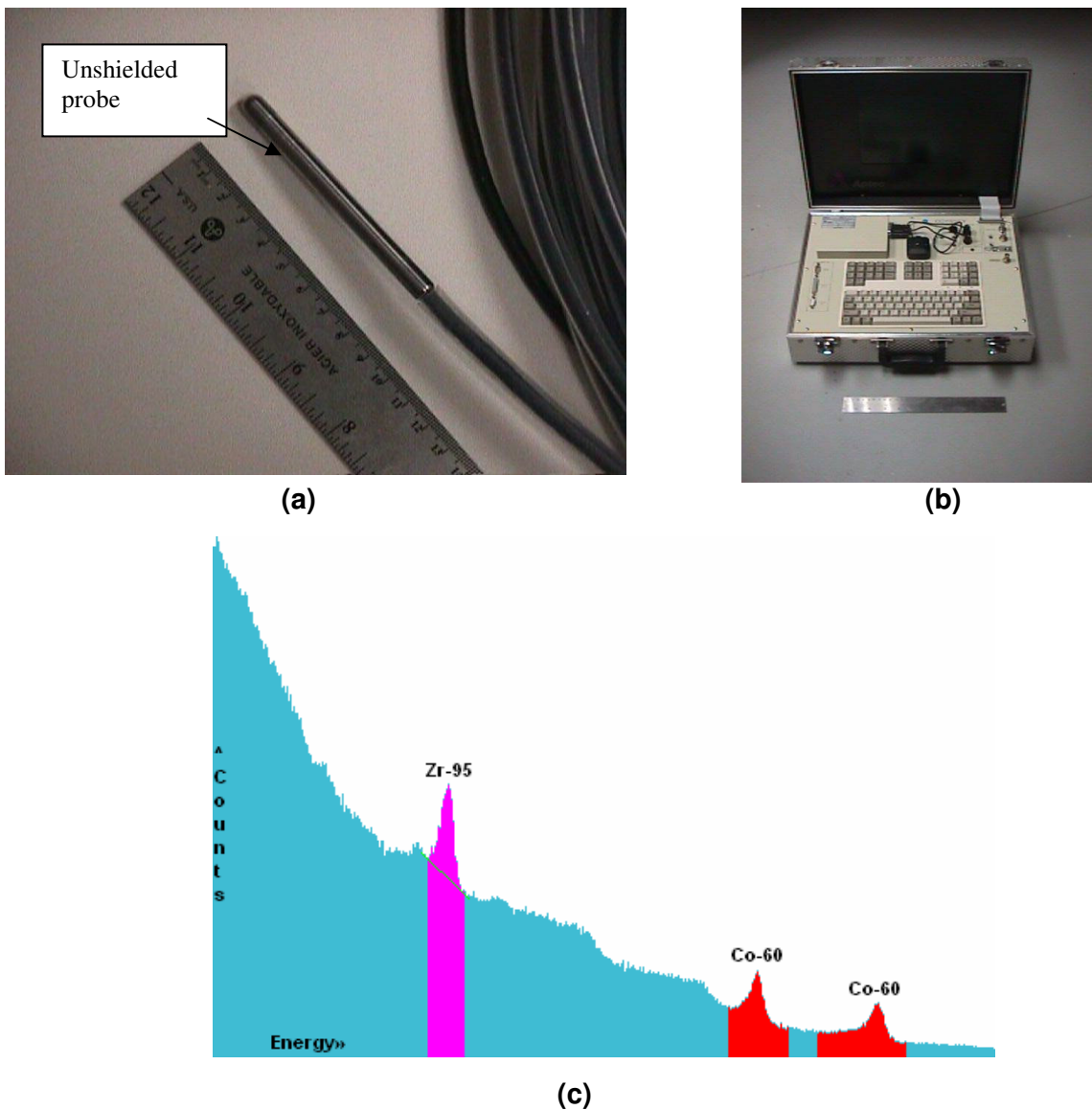


(d)

**Figure2**      **Photographs of the Filter Showing (a) & (b) End Views; (c) Filter Elements and d) Sampled Element**



While the above applications involved the use of germanium detectors, a unique application for ILW resin containers involved the use of cadmium-zinc-telluride detectors [Husain 1997]. These detectors are significantly smaller in size, have intermediate resolution compared with germanium and sodium iodide detectors and can be employed in radiation fields exceeding 1 Rem/h. Figure 3 shows a detector and a multichannel analyzer system along with the detector's response obtained in fields exceeding 1 Rem/h. Such a system was used to determine the activity of 3 m<sup>3</sup> spent resin containers which are stacked 6 high within an in-ground storage structure at the WWMF. An empty sample pipe runs parallel to the depth of the in-ground storage structure and is separated from it by a few inches of concrete and metal. The stored containers were scanned by lowering the gamma scan probe down the sample pipe to the required depth (middle of each stored container). Results obtained were consistent with those obtained from withdrawn samples of resin.



**Figure 3 (a) Cadmium Zinc Telluride Detector (b) APTEC's Multichannel Analyser (c) Gamma Spectra Obtained Using Cadmium-Zinc-Telluride Detector**

### 3.0 APPLICATION OF DOSE RATE TO ACTIVITY CONVERSION METHODOLOGY

Routine application of gamma spectrometry for assaying waste packages can be expensive. When the distribution of radionuclide activities is relatively constant for waste packages of a selected type, an alternate and simpler approach involves the use of Dose Rate-to-Activity Conversion (DRAC) methodology. The latter is based on the following consideration:

The dose rate  $D$  from gamma emitting nuclides in a waste package can be expressed as [Helmholz and Pyo 1983]

$$D = \sum (d_i C_i) \quad (1)$$

where  $d_i$  is the dose rate per unit activity or dose rate to activity conversion factor for a given nuclide  $i$  and  $C_i$  is the activity of the nuclide in the waste package. Because

$$C_i = f_i C \quad (2)$$

where  $f_i$  equals the fraction of nuclide  $i$  and  $C$  is the total activity, Equation (2) can be expressed as

$$C_i = f_i D / \sum (d_i f_i) \quad (3)$$

Thus, if  $d_i$  and  $f_i$  are known, the activity of individual radionuclides  $C_i$  and hence the total activity  $C$  can be determined from measured dose rates. The factor  $d_i$  is a function of density and can be determined from a shielding code while  $f_i$  is obtained from gamma scanning data. Dose rate measurements are typically performed at a measurement distance of 0.30 m (1 ft) at which there appears to be a reasonable compromise between the magnitude of the radiation response, the sensitivity of the detector to near surface distances for drums, and the sensitivity of the detector to non-uniform activity distribution at near surface distances.

Applicability of DRAC methodology was investigated for

- Drums containing assorted non-processible wastes,
- Boxes containing incinerator bottom ash
- Compacted waste boxes, and
- Boxes containing non-processible wastes.

In general, the population of each type of waste package investigated was relatively small. Although 100 drums were examined, they contained 10 different types of waste with fewer than 10 drums for several types. Similarly, the total number of boxes examined numbered only 20. The following conclusions were reached:

- For successful application of the DRAC methodology, the radionuclide distribution in different waste packages must be substantially constant and sufficient time must elapse after package generation to permit decay of short-lived radionuclides to insignificant levels. This duration in many cases is expected to be about a month.

- Co-60 activity in all the boxes examined was conservatively estimated with the measured/ estimated activity ratio being on average 0.71 (range 0.6-0.9). Cs-137 activity in most of the boxes examined was also conservatively estimated although the range in the measured/ estimated activity ratio was significantly wider. Despite variable results for individual boxes, the average of the measured/estimated activity ratios for Co-60, Cs-137, Zr-95, Nb-95 and Mn-54 were close to unity (0.71-1.1). This suggests that while activities for individual boxes may be over/under estimated, the inventory estimate for a large population of boxes may be reasonably accurate.
- The discrepancy between measured and estimated activities was most severe for cases where the actual activity distribution differed significantly from the average fractional activity distribution utilized to estimate the activity. Hence, the fractional activity distributions should be based on activity data for a larger number of packages.
- Application of the DRAC methodology will result in the apportioning of the total radioactivity in each package to all the radionuclides making up the fractional activity distribution. All the packages examined contained Co-60, most but not all contained Cs-137 and many of the packages did not contain one or more of the shorter-lived radionuclides. The attribution of Cs-137 and other radionuclides to a package, that in fact does not contain them, under-estimates the activity of Co-60 although the estimate may still be conservative.
- Application of the DRAC methodology to older packages would require the fractional activity distributions to be corrected for radioactive decay.

#### **4.0 EXPERIMENTAL DEVELOPMENT OF SCALING FACTORS**

Scaling factors are generally employed to estimate the concentrations of DTM radionuclides by relating their activities to those of easy-to-measure (ETM) gamma-emitting radionuclides. Nuclides with similar physical/chemical characteristics and produced/released in a similar manner can be expected to exhibit a strong correlation. Unfortunately, the choice of relatively long-lived ETM radionuclides is generally limited to the radionuclides Co-60, Cs-137, Sb-125, Ce-144 and Ru-106 with Co-60 and Cs-137 being frequently the only practical choices. Measured radionuclide activity data are typically correlated as follows:

- (Pu-239+Pu-240) is scaled relative to Co-60 and Cs-137. It may also be scaled relative to Ce-144, Sb-125 and Ru-106,
- The actinides Pu-238, Pu-241, Am-241, Cm-242 and Cm-244 are scaled relative to (Pu-239+Pu-240) (although these activity ratios do not conform to the definition of a scaling factor, they are, hereafter, for simplicity, also referred to as scaling factors),
- Scaling factors for C-14, Sr-90 and Tc-99 are determined relative to both Co-60 and Cs-137,
- Fe-55, Ni-63 and Cl-36 are scaled relative to Co-60 only.

- I-129 is scaled relative to Cs-137 only.

Note that scaling actinides relative to (Pu-239+Pu-240) allows effective use of sample actinide data even when gamma activity data for ETM radionuclides are not available.

#### 4.1 Scaling Factors for OPG Waste Streams and Data Treatment

Scaling factors are being developed [Husain 1995, 2001] for a number of LLW and ILW streams including:

LLW: incinerator bottom ash, incinerator baghouse ash, compacted waste, non-processible wastes, CAN-DECON resins, ALWTS resins, ALWTS sludges, Heavy Water Upgrader (HWU) charcoal, HWU resins, Irradiation Fuel Bay and other miscellaneous system filters.

ILW: Moderator System resins, Primary Heat Transport System (PHTS) resins, Moderator System filters and PHTS filters.

Scaling factor data are developed primarily based on samples collected from individual waste streams. Data based on smears collected from various plant areas have also been utilized. Although the development of data based on waste samples poses greater analytical challenges because of the diverse constituents of waste, it has the advantage of not introducing artifacts from the smearing process. Description of the technique utilized for sampling resins is provided in a companion paper.

Analysis of the large amount of scaling factor data is performed using the Sample Analysis Program (SAP) developed by David James & Associates, North Oaks, Minnesota. The program has the capability of generating scatter and trend plots for desired combinations of DTM and marker radionuclides; the scatter plots indicate the origin of each data point and also identify outliers. In addition to the data plots, SAP also generates a statistical report showing the geometric or log mean (LM) and log dispersion<sup>2</sup> (LD) values for the entire data set and for each waste stream included in the data set. A matrix of pooled variance factors<sup>3</sup> is also generated for each scaling factor.

Based on an analysis of pooled variance factors, it was concluded that scaling factors for bottom ash, including those for C-14, are essentially similar to those for various LLW waste streams (excluding baghouse ash). This observation suggests the feasibility of updating the LLW scaling factors database based solely on data for bottom ash. Note that bottom ash is easier to sample and analyse compared to the other LLW waste streams because of its particulate form and less variable composition.

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<sup>2</sup> LD is calculated by first taking the logarithms of individual data points, calculating the standard deviation of the logarithms and finally taking the inverse logarithm of the standard deviation. Note that a new measurement has a 68.3% probability of being within the range defined by the quantities LM x LD and LM/LD.

<sup>3</sup> SAP takes the data set for each stream individually and compares it to the data for other streams. The comparison determines the factor of difference between the data sets based on sample counts, variance and means. A value of 1 indicates that the data from the two sets are not distinguishable at the 95% confidence level and hence segregating the waste streams is not called for. A value of 2 indicates a factor of 2 difference.



## 4.2 Estimation of Tritium Concentrations

Direct measurement of tritium concentrations in bulk LLW presents a very significant challenge. The feasibility of determining the tritium inventory in waste packages has been assessed at a laboratory scale. Further development is, however, required.

Attempts to scale H-3 concentrations relative to Co-60 have generally met with limited success. The high mobility of tritium is a significant impediment in the reliable quantification of tritium present in waste samples.

Despite the differences in their chemical behaviour, a correlation may be expected between C-14 and H-3 because their dominant production mechanisms, namely  $O-17(n,\alpha)C-14$  and  $H-2(n,\gamma)H-3$ , involve the activation of heavy water constituents. The availability of tritium and C-14 emissions data for the radioactive waste incinerator at the WWMF offers the possibility of correlating the concentrations of H-3 and C-14 present in bagged LLW (compacted and non-processible wastes) based on their relative concentrations in large volumes of incinerable waste. This would allow H-3 concentrations to be determined from C-14 concentrations with the latter, in turn, being determined from the C-14/Co-60 scaling factor.

The variation of the C-14/H-3 activity ratio based on weekly incinerator emissions and burn data for 2003 is illustrated in Figure 4. Most of the data values range between  $1E-04$  and  $1E-05$ . The average value for the entire duration considered (the volume of waste incinerated during this period was  $\sim 1500 \text{ m}^3$ ) was  $6.2E-05$ . For reference, the C-14/H-3 ratio in the Pickering-A moderator coolant is approximately  $10^{-7}$  (unfortunately, the corresponding value for the PHT coolant is not known).

The C-14/H-3 value of  $6.2E-05$  is the present basis for estimating the H-3 concentrations in OPG's bagged waste.

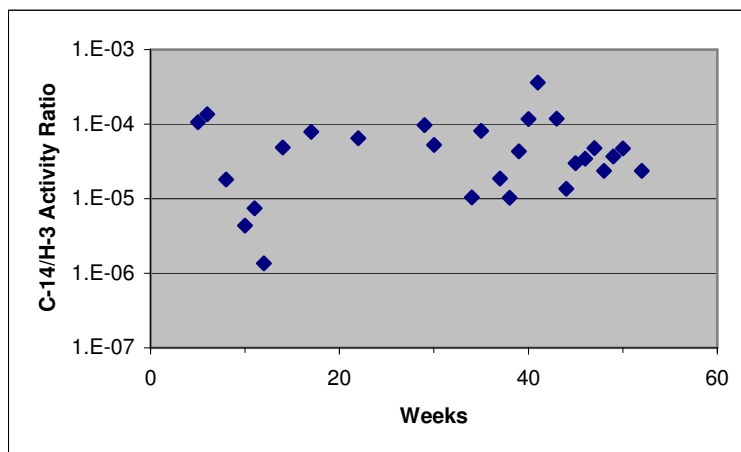


Figure 4. Variation of the C-14/H-3 Activity Ratio in Incinerable Waste in 2003

## 5.0 PREDICTIVE MODELS FOR ESTIMATING SCALING FACTORS

Methods for predicting scaling factors depend on whether the radionuclides of interest are generated by fission or by neutron activation. The following approaches have been employed:

- Predictions based on fission product release models,
- Predictions based on activation models, and
- Predictions based on estimated radionuclide inventories in used fuel.

Predictive capability is being developed in order to reduce the need for developing an extensive scaling factor database. Amongst the various DTM radionuclides, I-129, Tc-99 and Se-79 present a greater analytical challenge because of their much lower concentrations in reactor waste.

### 5.1 Predictions Based on Fission Product Release Models

Fission product release models are based on a description of the release process of fission products from defective fuel rods and from uranium contamination present on in-core reactor surfaces. The release of any chemical element from defective fuel is dictated by its ability to diffuse through the grain ( $R_d$ ) and then vaporize from the grain surface ( $R_v$ ) where  $R$  denotes the release rate. The overall release rate  $R$  equals the minimum of  $R_d$  and  $R_v$ . If  $R_v \gg R_d$ , the rate determining step is diffusion in the fuel matrix. Mobile species, which accumulate in the fuel-clad gap are transported from the gap into the coolant via the defect site. This second transport step has been postulated to be a first order rate process. In contrast to the release processes from defective fuel, fission products are released directly by recoil from in-core tramp uranium contamination.

A mass balance, considering the two fuel source terms (defective fuel and tramp uranium contamination) and loss terms due to coolant cleanup and radionuclide decay were solved to estimate fission product activity in the PHT coolant. Steady state assumptions are made to solve for the activity of short-lived fission products. Solutions for long-lived fission products are time-dependent. Coolant data for short-lived radioiodines I-131, I-132, I-133, I-134 and I-135 and/or noble gases Kr-85m, Kr-87, Kr-88, Xe-133m, Xe-135 and Xe-138 are required to estimate model parameters. The estimated parameters can then be used to determine the activity of long-lived DTM radionuclides present in the coolant.

To-date, fission product release models have been developed for predicting the scaling factors for I-129, Tc-99 and Se-79 [Lewis et al 2004a,b; Lewis and Husain 2003; Lewis et al 2003; Lewis and Thompson 2003]. These models have distinctive features arising from the specific behaviour of each fission product under reactor operating conditions.

Predicted results based on radiochemical data for the Darlington PHTS coolant are summarized in Table 1. The values for I-129 and Tc-99 scaling factors are consistent with data reported for PWRs and BWRs and with the limited data presently available for OPG's ILW. No OPG data are yet available for Se-79.

**Table 1 Predicted Values for Scaling Factors of Fission Product Radionuclides**

Scaling Factor	Predicted Value
I-129/Cs-137	1E-08-1E-07
Tc-99/Cs-137	1E-06
Se-79/Cs-137	3.6E-08

Note that values of 2.9E-07 and 1.3E-04 were, respectively, estimated for I-129/Cs-137 and Tc-99/Cs-137 scaling factors based on radionuclide inventory tabulations for used fuel (see Section 5.3). The estimated value for Se-79/Cs-137 is roughly an order of magnitude smaller than that estimated from the radionuclide inventories in used fuel.

## 5.2 Predictions Based on Activation Models

The DTM radionuclides C-14, Cl-36, Ni-63 and Fe-55 and the ETM radionuclide Co-60 are all produced by neutron activation in the reactor core. Therefore, the scaling factor for each DTM radionuclide, relative to Co-60, can be estimated from neutron activation considerations. The relative ratio of the activities ( $A_1$  and  $A_2$ ) of two activation radionuclides is given by

$$A_2 = \frac{\sigma_2 \lambda_2 M_2}{\sigma_1 \lambda_1 M_1} A_1 \quad (4)$$

where:

$\lambda$  is the radioactive decay constant ( $s^{-1}$ ),

$M$  is the moles of inactive element in-core, and

$\sigma$  is the thermal absorption cross-section for the inactive element (barns).

Equation (4) was applied to estimate values for the various scaling factors considering PHT system conditions. The  $M_2/M_1$  ratios in these cases were obtained as follows:

- Because C-14 is produced by the  $(n, \alpha)$  reaction of O-17, the in-core inventory of O-17 target nuclides was estimated from the concentration of O-17 in and the in-core volume of the PHT heavy water.
- Cl-36 is produced principally by the  $(n, \gamma)$  reaction of Cl-35. The several sources of chlorine in the PHT and moderator systems lead to ppb levels of chlorine in both coolant systems. Therefore, the in-core inventory of Cl-35 in the PHT system can be estimated similarly to the case of O-17; the corresponding estimate for the moderator system is obtained based on the total mass of the moderator coolant.
- Fe-55, Ni-63 and Co-60 are produced by the  $(n, \gamma)$  reaction of Fe-54, Ni-62 and Co-59, respectively. The in-core inventory of the target nuclides have to-date been estimated only for the Pickering PHT system (no data were available for Bruce and Darlington PHT systems and none for moderator systems in general). The estimation for the Pickering PHT system was based on the mass of corrosion product deposits on pressure tube and fuel bundle surfaces and their associated Co/Fe and Ni/Co ratios [Burrill 1996]. Because of the large variability in these data, the estimates are subject to significant uncertainty.

The estimated values for C-14/Co-60, namely, 0.12 is consistent with the overall LM value of 0.064 as shown in Table 2. The estimated value for Cl-36/Co-60 in the PHT system, namely, 1.1E-06, is a factor of ~10 lower than the LM value of 1.0E-05 measured for moderator resins.

**Table 2 Predicted Values for Scaling Factors of Activation Radionuclides**

Scaling Factor	Estimated Value	Measured Value*
C-14/Co-60	0.12	LM=0.064, LD = 8.8
Cl-36/Co-60	1.1E-06	LM=1.0E-05, LD=2.2
Ni-63/Co-60	0.25	LM= 2.4E-02, LD=6.2
Fe-55/Co-60	0.60-75	LM=3.4, LD=4.2

\* Measured values except in the case of Cl-36/Co-60 are for LLW; the value for Cl-36/Co-60 is based on moderator resin data.

### 5.3 Predictions Based on Specific Activities of Radionuclides in Used Fuel

Specific activities for various radionuclides present in irradiated fuel have been estimated as a function of fuel burnup and post-irradiation decay time,  $t_{p-irr}$  [Tait et al 1989]. The ratios of DTM radionuclides to selected ETM radionuclides, based on these compilations may be reasonable approximations for scaling factors when the release behaviour (from fuel) of the pertinent radionuclides and their chemical and solubility behaviours in the coolant are similar.

Activity ratios for a typical burnup of 180-190 MWh/kg and  $t_{p-irr}$  of 1 year are shown in Table 3 [Husain and Krasznai 1995]. With the exception of the ratios of Cm-242/Pu-239 and Am-241/Pu-239, the activity ratios vary exponentially with decay time as  $\exp(\lambda t_{p-irr})$  where  $\lambda$  is the effective rate constant. The variation of Cm-242/Pu-239 and Am-241/Pu-239 with  $t_{p-irr}$  is much more complex: the two ratios decrease and increase, respectively, with  $t_{p-irr}$  before eventually leveling out. The activity ratios, in general, also depend on burnup. This dependence is relatively small for the activity ratios Pu-239/Ce-144, Sr-90/Cs-137, Tc-99/Cs-137, I-129/Cs-137, Pu-240/Pu-239 and Pu-241/Pu-239.

The effective values for fuel burnup and  $t_{p-irr}$ , which correspond to the measured radioactive waste characteristics, can be determined as follows:

- The ratio Cm-244/(Pu-239+Pu-240) has a relatively strong dependence on burnup but a small dependence on  $t_{p-irr}$  ( $\lambda = -0.038 \text{ y}^{-1}$ ). Therefore, the measured ratio can be utilised to estimate burnup from the used fuel specific activity compilations.
- Based on the estimated burnup, values for Cm-242/(Pu-239+Pu-240), Am-241/(Pu-239+ Pu-240) or Pu-241/Pu-239+Pu-240) at  $t_{p-irr} = 0$  can be determined from the activity compilations for used fuel. The applicable value for  $t_{p-irr}$  can then be determined from the estimated ratios at  $t_{p-irr} = 0$  and the measured ratios.

Using the values for burnup and  $t_{p-irr}$  thus determined, the used fuel tabulations can be utilised to estimate other specific activity ratios of interest.

**Table 3 Estimated Activity Ratios for Various Radionuclides Based on Their Specific Activities in CANDU Fuel**

<b>Nuclide Ratio</b>	<b>Pickering Fuel Burnup 181 MWh/kg U <math>t_{p-irr} = 1 \text{ y}</math></b>	<b>Bruce Fuel Burnup 190 MWh/kg U <math>t_{p-irr} = 1 \text{ y}</math></b>	<b><math>\lambda \text{ (y}^{-1}\text{)}</math></b>
Pu-239/Ru-106	0.0016	0.0016	0.69
Pu-239/Cs-137	0.0067	0.0067	0.023
Pu-239/Ce-144	8.9E-04	7.4E-04	0.89
Sr-90/Cs-137	0.67	0.67	-1.1E-03
Tc-99/Cs-137	1.4E-04	1.4E-04	0.023
I-129/Cs-137	2.9E-07	3.1E-07	0.023
Cm-244/Pu-239	0.07	0.099	-0.038
Cm-243/Pu-239	0.0015	0.001	-0.024
Cm-242/Pu-239	2.7	1.6	-
Am-241/Pu-239	0.23	0.24	-
Pu-241/Pu-239	110	130	-0.047
Pu-240/Pu-239	1.4	1.4	-2.2E-04
Pu-238/Pu-239	0.44	0.5	-7.0E-03

## 6.0 CLOSING REMARKS

Kinectrics has been involved in the radiochemical characterization of OPG's low and intermediate level waste since the early 80's. Until about 1995, characterization had largely been limited to the measurement of gamma activity based on the assay of waste samples or waste bags. Since then, capability has been developed for the gamma assay of large waste packages. Also, scaling factors have been developed for numerous waste streams.

Because of the need for a relatively large dataset (for statistical significance) and the high cost of quantifying  $\alpha$  and  $\beta$  emitters, significant expense is associated with the development of scaling factors. Since 1999, scaling factors for several DTM radionuclides present in OPG's LLW have been annually updated based on newly developed experimental data. In general, sufficient scaling factor data now exist for all bulk LLW waste streams except non-processible wastes. This data pertains to the DTM radionuclides C-14, Fe-55, Ni-63, Sr-90, Pu-238, (Pu-239+ Pu-240), Pu-241, Am-241, Cm-242 and Cm-244. In contrast, the scaling factor data for OPG's ILW are very limited and restricted to PHT and moderator resins. New resin sampling techniques have enabled the development of data for ILW moderator resins; sampling of PHT resins is planned in 2005.

ILW resins need to be characterized also for I-129, Tc-99, Cl-36 and Se-79. This need has, therefore, led to the development of analytical protocols for their measurement. In addition, the desirability of limiting the size of the dataset for these radionuclides (the measurements are time consuming and dose intensive), has led to the development of models for the release of I-129, Tc-99 and Se-79 from fuel and for Cl-36 based on activation considerations. The fission product models are capable of estimating not only



the desired scaling factors but also the cumulative releases of these radionuclides over several years of reactor operation.

The gamma activity and scaling factor data generated to date have been used to compile radionuclide inventory data for OPG's currently stored waste. In instances where data does not presently exist, data for other similar waste streams are utilized. These inventory compilations are annually updated as new data become available. Thus, OPG has gained a reasonable and defensible radiochemical database and a good understanding of the characteristics of their waste. This will be invaluable as it moves forward with the repository project.

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