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# ESTIMATION OF THE RADIONUCLIDE INVENTORY OF ACTIVATED DECOMMISSIONING WASTE FROM ONTARIO POWER GENERATIONS'S DARLINGTON NUCLEAR GENERATING STATION

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## ABSTRACT

This paper presents the results of a detailed assessment of the neutron activated radionuclide inventory in decommissioning waste from Darlington.

A PC-based code was used to estimate the radionuclide concentrations in activated reactor core components. For each component, the inputs included a) power history, b) material composition data, c) applicable value of neutron flux d) a thermal flux correction factor, e) component mass and f) temperature. Various OPG documents were examined to assemble the required data inputs.

The overall activity, 33 years after shutdown, for each Darlington unit was estimated to be 4.2E+16 Bq. Ni-63 accounts for approximately 95 % of this activity. Other key radionuclides are Co-60, Ni-59, Nb-94, C-14, Fe-55, Zr-93, Mo-93 and Cl-36. The end shields are the dominant source for C-14, Fe-55, Co-60, Ni-59, Ni-63 and Cl-36 activities. The pressure tubes and the calandria tubes are the principal sources for Zr-93. Nb-94 is associated exclusively with pressure tubes.

Estimated radionuclide activities were generally consistent with previous estimates for Pickering-A and Bruce-A stations.

## 1.0 INTRODUCTION AND BACKGROUND

Ontario Power Generation (OPG) is a government owned electrical utility operating in the province of Ontario, Canada. It owns five 4-Unit Nuclear Generating Stations (NGS). These are located at three sites, namely, Pickering (Stations A and B), Bruce (Stations A and B) and Darlington. Although all the plants are based on the CANDU reactor concept, key differences, exist between their designs such as unit power, plant layout and materials of construction.

For long term planning purposes, it is assumed that OPG stations will be shut down following their 40-year operating lives and decommissioned in order of their in-service dates. The assumed shutdown date for the last unit at Darlington is 2033. A delayed dismantling strategy is assumed for decommissioning the nuclear stations. The strategy is characterized by the following three stages:

- Stage 1 - Preparation for Safe Storage: This involves the removal of used fuel from the reactor core and chemical decontamination of the primary heat transport (PHT), moderator, and other radioactive systems.

- Stage 2 - Safe Storage: The 30 years duration of Stage 2 permits significant decay of radioactivity to occur, thus minimizing worker dose during the next decommissioning stage. An environmental surveillance is maintained to control potential radioactivity releases from the plant.

- Stage 3 - Preparation for Dismantling, Dismantling and Site Restoration: The Stage 3 dismantling phase can last up to 10 years. Site buildings are decontaminated before being dismantled. Conventional systems are also dismantled.

A description of the waste arisings from each OPG station along with their volumes were developed by TLG Services Inc. as part of an overall study to estimate decommissioning costs. A detailed analysis of the TLG waste arisings data was subsequently performed by Kinectrics. In addition, a separate study focussed on the estimation of radionuclide inventories associated with Darlington's activated decommissioning waste. Results from this study complement the radionuclide inventory data developed for Pickering and Bruce NGSs in earlier Ontario Hydro studies [1986a,b]. This paper presents an overview of the radionuclide inventory assessment for Darlington.

## 2.0 OVERVIEW OF THE REACTOR SYSTEM AT DARLINGTON NGS

Figure 1 shows a schematic for a single unit CANDU Pressurised Heavy Water plant. The four 881 MWe (net) units at Darlington are physically interconnected and share a common vacuum building (this building serves to relieve over-pressurisation during an accident situation). Each unit is comprised of a reactor building supported by a reactor auxiliary bay and the powerhouse; the latter is one large unit housing the turbine/generator sets for all 4 units.

The reactor building is divided vertically into 3 areas: the fueling duct and basement, the reactor vault and rooms above the reactor vault which house the steam generators, the heat transport pump motors and reactivity drive mechanisms.

The reactor vault (see Figure 2) forms part of the containment envelop. The shield tank assembly serves as the biological shielding around the reactor. It extends from the floor to the celing of the vault. The top surface of the shield tank extension supports the reactivity mechanism deck.

The reactor assembly is illustrated in Figure 3 and comprises 1) the calandria and shield tank assembly, 2) the fuel channel assemblies with integral end fittings and 3) the reactivity control units. The calandria and shield tank assembly consists of:

- the horizontal, cylindrical, calandria vessel spanned axially by 480 calandria tubes; the calandria tubes contain and support the fuel channels or pressure tubes and isolate them from direct contact with the heavy water moderator contained in the calandria.

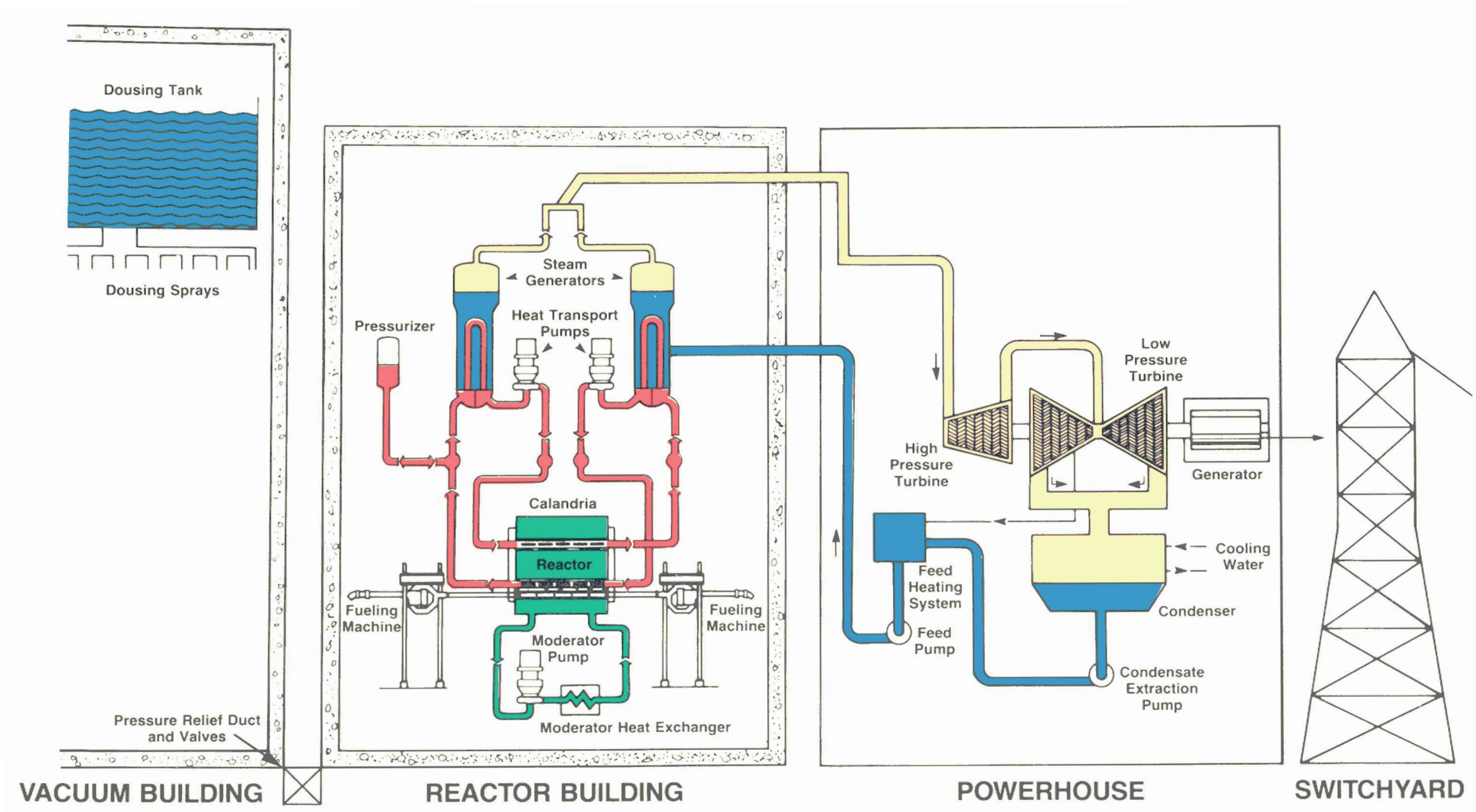



**Figure 1 Schematic of CANDU Pressurised Heavy Water Reactor Unit**

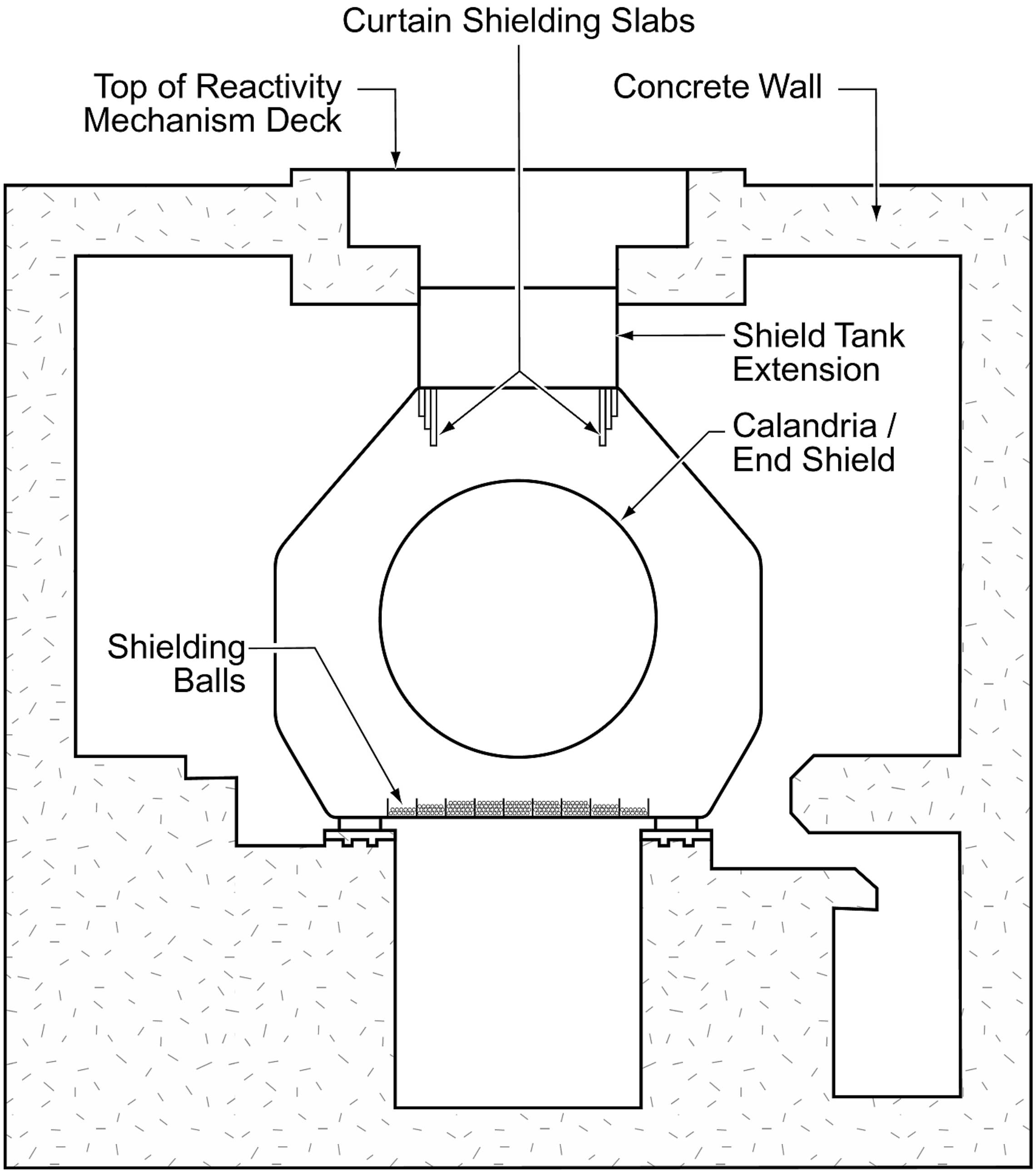
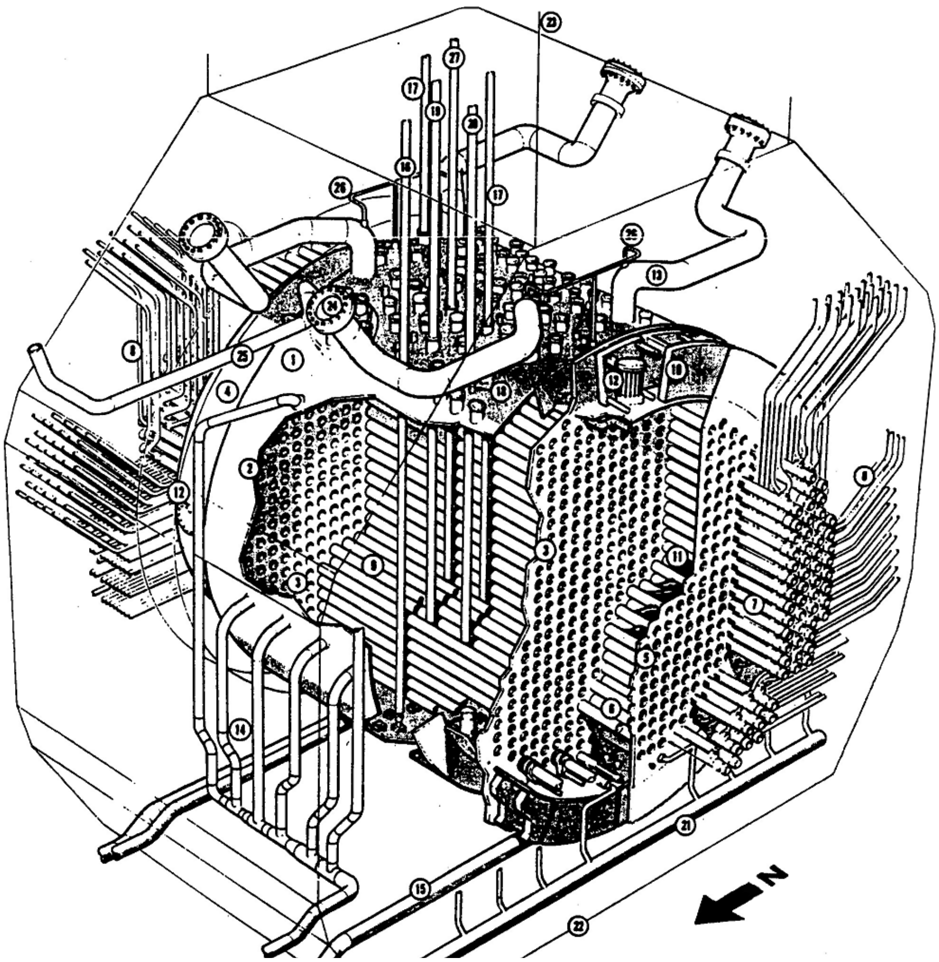



**Figure 2** **A Cross-Section Across the Reactor Vault** 2000064 10 01



200060 10 01
8958 050

1. Calandria
2. Calandria Main Shell
3. Calandria Side Tubesheet
4. Calandria Sub Shell
5. Fuelling Machine Side Tubesheet
6. Lattice Tubes
7. End Fittings
8. Feeders
9. Calandria Tubes
10. Shield Tank Solid Shielding
11. Steel Ball Shielding (End Shield)
12. Manhole
13. Emergency Discharge Pipes
14. Moderator Inlets
15. Moderator Outlets
16. Shut-off Unit
17. Adjuster unit
18. Vertical Flux Detector
19. Control Absorber
20. Liquid Zone Control Unit
21. End Shield Cooling Piping
22. Shield Tank
23. Shield Tank Extension
24. Rupture Disc Assembly
25. Moderator Overflow
26. Pressure Balance Lines
27. Viewing Port

**Figure 3 Darlington NGS Reactor Assembly**

- two integral, horizontal, cylindrical, end shields spanned horizontally by 480 lattice tubes; the end shields contain shielding material in the form of carbon steel balls and light water, and

- the shield tank - this is a carbon steel vessel with double end walls and an upper rectangular extension. The calandria vessel and the end shields are installed in the shield tank. The shield tank also contains light water, steel shielding slabs and shielding balls.

The reactivity mechanism deck closes the top of the shield tank to provide a containment boundary between the light water in the shield tank and the accessible area above the deck. It provides a mounting area for the vertical reactivity control units.

The vertical reactivity mechanisms consist of liquid zone control units, adjuster units, shutoff units, solid control absorber units and vertical flux detector units. Three types of horizontal reactivity control units penetrate the shield tank, namely, ion chambers, liquid injection shutdown units and horizontal flux detectors.

## 3.0 SCOPE OF CALCULATIONS

Table 1 presents a breakdown of the various reactor components and their assumed classification as decommissioned reactor waste.

Estimates of radionuclide activities associated with the various components listed in Table 1 were developed considering only neutron-induced activation. Activity from deposited activated corrosion products is implicitly accounted for in these estimates. Compared with the activities of activated radionuclides, those of fission products deposited on system surfaces are expected to be relatively small. Radionuclides deposited on system surfaces, in general, are likely to be partially removed from the reactor system as a result of decontaminations performed during the operating life of the reactor. Thus, the inventory of fission products in decommissioning waste would be further reduced.

The overall scope of the present calculations are summarised in Table 2. Because the calandria vessel is completely surrounded by the shield tank, neutron fluxes beyond the shield tank wall are insignificant and result in negligible activation of the reactor vault concrete wall.

## 4.0 CODE USED FOR ESTIMATING NEUTRON-INDUCED ACTIVITIES AND DATA INPUTS

A Simplified Neutron Activation Analysis Program (SNAP), recently developed by D.W. James & Associates [James 2001], was used for estimating neutron-induced activities. SNAP is a Windows adaptation of the more complex ORIGEN code which has been widely used for neutron activation analysis. It is written in $C^{++}$. The program inputs and the methodology employed to develop them are discussed below:

## Table 1 Darlington NGS Reactor Components - Mass and Waste Classification

| Component | Quantity Per Unit | Mass (lb) per Component | Type of Waste* |
|---|---|---|---|
| **Shield Tank-Calandria-End Shields Assembly** | | | |
| Shield Tank (less extension) | 1 | 689,000 | LLW |
| Calandria vessel | 1 | 83,990 | LLW |
| Calandria tubes with inserts | 480 | 54 | **ILW** |
| Internal piping (shield tank to calandria) | 1 | 15,800 | LLW |
| End Shields | 2 | 169,570 | LLW |
| **Shield Tank Extension** | 1 | 69,970 | LLW |
| **Steel Balls** | | | |
| Shield Tank | 1 | 180,500 | LLW |
| Shield tank extension | 1 | 142,700 | LLW |
| End shields | 2 | 283,400 | **ILW** |
| **Solid Shielding – Shield Tank** | | | |
| Curtain shielding slabs | 2 | 27,531 | LLW |
| Annular shielding assemblies | 72 | 2,820 | LLW |
| **Fuel Channels** | | | |
| End fitting assemblies | 960 | 379 | **ILW** |
| Closure plugs | 960 | 28 | LLW |
| Shield plugs | 960 | 173 | **ILW** |
| Liner and latch | 960 | 70 | LLW |
| Pressure tubes | 480 | 138 | **ILW** |
| Grayloc fittings | 960 | 18.9 | LLW |
| **Reactivity Mechanism Deck** | | | |
| Deck structure | 1 | 127,166 | LLW |
| Vertical shielding | 1 | 43,206 | LLW |
| Concrete shielding | 1 | 152,895 | LLW |
| Tread plates and floor plates | 1 | 59,527 | LLW |
| Manhole plug | 1 | 1,174 | LLW |
| Shielding collars | 1 | 16,897 | LLW |
| Zone Control Unit (ZCU) shielding box and support | 2 | 22,687 | LLW |
| ZCU[b] horizontal shielding slab | 1 | 21,466 | LLW |
| **Other** | | | |
| Control rods with drives and ion chambers | 1 | 128,000 | **ILW** |
| Feeder tubes | 1 | 388,875 | LLW |
| External piping - end shield cooling (full) | 2 | 6,500 | LLW |
| - shield tank cooling | 1 | 2,000 | LLW |
| Feeder tube supports and hangars | 1 | 8,600 | LLW |
| Feeder cabinet | 2 | 2,000 | LLW |
| Gap shielding tank extension to concrete | 1 | 37,000 | LLW |
| Shield tank support bearings incl. base plate | 4 | 16,400 | LLW<br>LLW |

* LLW, Low Level Waste; ILW, Intermediate Level Waste

## Table 2 Overall Scope of Neutron Activation Calculations

| Component (see Table 1) | Scope of Calculations |
|---|---|
| Shield Tank-Calandria-End Shields Assembly | All components except the shield tank were included in the scope of calculations. The neutron flux at the shield tank wall is considered to be too low to cause significant activation. |
| Shield Tank Extension | Similar to the shield tank wall, the vertical shield tank extension is expected to undergo negligible neutron induced activation. |
| Steel balls | The steel balls in the shield tank extension were disregarded. |
| Solid Shielding - Shield Tank | Activation of both the curtain and annular shielding assemblies was considered. Their applicable neutron flux values were, however, not well defined. |
| Fuel Channels | Except pressure tubes, components listed under 'Fuel Channels' (see Table 1) consist of several sub-components, each with varying materials of construction. Calculations were done separately for individual sub-components and then summed to obtain the inventory for the overall component. |
| Reactivity Mechanism Deck | Activation of the components of the reactivity mechanism deck arises only from neutrons streaming up the reactivity control unit penetrations. This will cause localized activation around the penetrations but is unlikely to cause bulk activation of the deck materials. Hence these components were excluded from the scope of calculations. |
| Other | The reactivity control mechanisms and feeder pipes are considered to be the only significant contributors in this category. The mechanisms contain up to 10 different sub-components. Calculations were done separately for individual sub-components and then summed to obtain the inventory for the overall component. Feeder pipes were excluded from the scope because the activation contribution to their activity is likely to be relatively small. |

Power History: The power history, maintained through a file, includes start/end dates and the corresponding % power level for each time period. Based on data for the various Darlington NGS units to date, a conservative value of 85% full power was selected over the entire operating duration of each unit.

Neutron Flux: Available data from various OPG sources were used to compile neutron flux values for components of interest. A series of graphs depicting the variation of the fast, intermediate and thermal neutron flux values as a function of distance across the end shield and across the thickness of water in the shield tank were utilised. The quality of the data ascertained from these plots was considered adequate for developing 'ball park' estimates (even with accurate flux information, dose rate measurements are typically required to fine tune activity estimates).

Because of the significant variation in flux values with distance, average values were estimated, where appropriate, assuming an exponential distance dependence. The distance averaged flux is accordingly given by the expression $(\varphi_2 - \varphi_1) / \ln(\varphi_2/\varphi_1)$ where $\varphi_1$ and $\varphi_2$ correspond to values of the flux at the two ends of the component.

Material Compositions of Reactor Components: Specification of each material includes its density and its detailed chemical composition**.** The latter was compiled (up to 25 elemental constituents) based on the following approaches, recognising that uncertainty in levels of minor constituents, such as cobalt, may have a significant impact on estimated radionuclide inventories:

- Materials certification data was sought from selected vendors.
- Materials certification data for several components were extracted from history dockets maintained at Darlington.
- Where the above approaches were unsuccessful, generic material compositions were obtained from various literature sources.

Mass of Various Reactor Components: The masses of various reactor components are listed in Table 1. Because the components, in several cases, consist of sub-components with assorted materials of construction, a further break down of the mass was obtained based on dimensions and density.

Thermal Flux Correction Factor: Spectrum averaged cross-sections were used in conjunction with flux values for in-core components. For out-core components, a correction factor was used to account for the higher fraction of thermalized neutrons. It has a user specified value between 0 and 1 and automatically adjusts the spectrum averaged cross-sections for additional thermal contribution.

The correction factor was estimated from the formula $r' = (r_2-r_1)/(1-r_1)$ where $r_1$ denotes the thermal to total flux ratio in the core energy-averaged cross sections and $r_2$ is the corresponding ratio in the vicinity of the irradiated object. Use of this formula requires that $r_2$ cannot be less than $r_1$. For CANDU plants, $r_1$ is approximately 0.2. Values of $r_2$ for various components were assembled from the applicable values of fast, intermediate and thermal fluxes.

Thermal Correction Temperature: This is used to correct thermal cross-sections data, which generally have a reference temperature of 20ºC. The cross-sections ($\sigma$) at any desired temperature T(K) are related to the cross-sections at 20°C or 293°K according to the relationship $\sigma (T) = \sigma (293) (293/T)^{0.5}$. Thus, in the case of the calandria vessel for example, where the inlet and outlet temperatures are 35°C and 65°C respectively, the appropriate temperature for computation was considered to be 35°C because this yields a conservative estimate for the induced activity.

Decay Period: Radioactive waste inventories were estimated for a decay period of up to 50 years although two specific time periods following shutdown, namely 2.5 years and 33 years were of specific interest. The latter pertain, respectively, to the start of Stage 1 and Stage 3 decommissioning.

## 5.0 RESULTS OF SAMPLE CALCULATIONS

Based on the methodology described in Section 4, a detailed compilation of input data was developed for the various components listed in Table 1 and for their associated sub-components.

Computations were performed to estimate the induced activity in sub-components and hence in each overall component. Tabular data were compiled only for radionuclides with half-life exceeding 0.5 years. Data for the key long-lived radionuclides were graphed for decay periods up to 50 years after shutdown.

For purposes of illustration, results for the SS-304L calandria vessel are presented in Table 3 and Figure 4. As shown, the total activity decreases significantly with decay of Co-60 and then levels off approximately 30 years following shutdown. The activity thereafter is dominated by Ni-63 with much smaller contributions from Ni-59 and C-14.

**Table 3 Estimated Radionuclide Concentrations in SS 304 L Calandria Vessel**

| Radionuclide | Half-Life (years) | Activity (Ci/kg) | |
|---|---|---|---|
| | | 2.5 y after Shutdown | 33 y after Shutdown |
| Zn-65 | 6.69E-01 | 3.11E-06 | 5.91E-20 |
| Mn-54 | 8.56E-01 | 2.38E-02 | 4.50E-13 |
| Fe-55 | 2.60E+00 | 4.56E+01 | 1.34E-02 |
| Co-60 | 5.26E+00 | 8.73E+00 | 1.57E-01 |
| H-3 | 1.24E+01 | 6.42E-09 | 1.17E-09 |
| Ni-63 | 9.20E+01 | 1.05E+01 | 8.34E+00 |
| Si-32 | 6.50E+02 | 3.26E-11 | 3.16E-11 |
| C-14 | 5.74E+03 | 2.08E-02 | 2.07E-02 |
| Ni-59 | 8.00E+04 | 8.10E-02 | 8.10E-02 |
| Cl-36 | 3.01E+05 | 4.05E-09 | 4.05E-09 |
| Be-10 | 1.60E+06 | 3.28E-11 | 3.28E-11 |
| | **Total** | **6.50E+01** | **8.62E+00** |

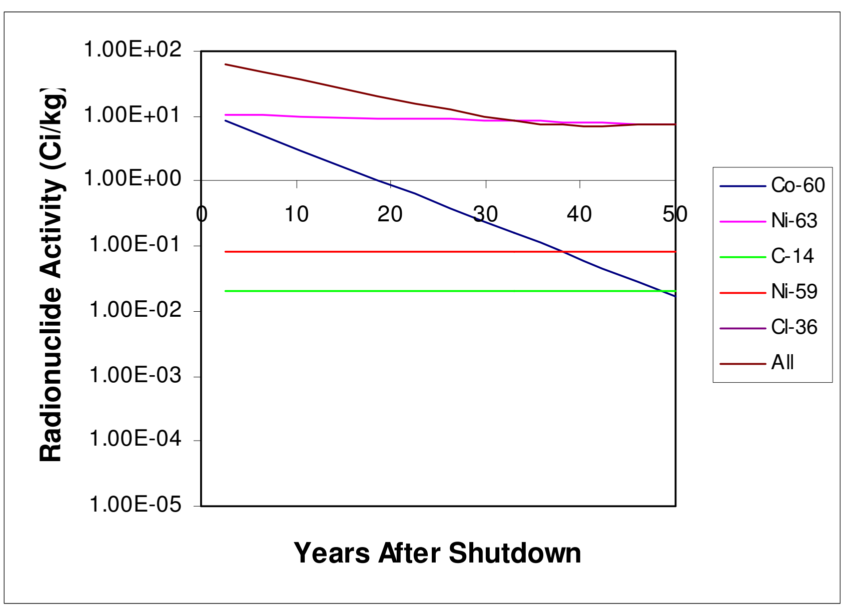



**Figure 4 Estimated Radionuclide Concentrations in SS 304 L Calandria Vessel**

It was desirable to compare estimates of induced activities in various components with the corresponding measured values. The latter, however, were available only in the case of pressure tubes [Chen et al 1993; Moir et al 1994]. The comparison is shown in Table 4. A slightly lower neutron flux value of 8.0E+13 n/cm$^2$/s (instead of 1.1E+14 n/cm$^2$/s) yielded 100% agreement with the measured activity data for Nb-94 and also Mn-54. Based on this flux value, the estimated and measured C-14 values agreed within 10% when the nitrogen content was increased from 38 to 42 ppm. Because no attempt was made to optimise other input parameters, Co-60 was over-estimated (the discrepancy may be partly attributable to the uncertainty in the decay history of the samples) while tritium was significantly under-estimated (the discrepancy is partly attributable to tritium ingress from the heavy water coolant into the pressure tube material during reactor operation).

**Table 4 Comparison Between Measured and Estimated Activities for Pickering Zr-2.5% Nb Pressure Tubes**

| Radio-Nuclide | Specific Activity Based on Data of Chen et al [1993] | | Specific Activity Based on Data of Moir et al [1994] | | Estimate Based on SNAP | Ratio of Estimated to Measured Activity |
|---|---|---|---|---|---|---|
| | LM* (Bq/g) | LD** | LM* (Bq/g) | LD** | (Bq/g) | |
| Co-60 | 2.4E+06 | 1.6 | 2.2E+06 | 2.8 | 7.6E+07 | 3.4E+01 |
| Mn-54 | - | - | 3.8E+04 | 3.7 | 3.8E+04 | 1.0 |
| Nb-94 | 5.3E+06 | 1.4 | 3.7E+06 | 2.7 | 3.8E+06 | 1.0 |
| H-3 | 1.0E+06 | 2.9 | 1.0E+06 | 3.0 | 2.7E+02 | 2.7E-04 |
| C-14 | 2.8E+05 | 1.7 | 1.9E+05 | 3.3 | 2.1E+05 | 1.1 |

* LM denotes log mean or geometric mean; ** LD denotes log dispersion

## 6.0 DEALING WITH UNCERTAINTY IN METAL COMPOSITIONS

Metal compositions for CANDU core components fall into 3 main categories, namely, steel alloys, nickel alloys and zirconium alloys. Within each category, the effect of the detailed metal composition on estimated activity concentrations was assessed for the following reasons:

- Metal composition data were not readily available for each component or sub-component of interest. In some cases, even generic compositions could not be obtained from handbooks. Assessing the computed activity concentrations as a function of various metal compositions provided guidance when the detailed composition was not known.

- The estimations also provided guidance on the need for detailed material characterization.

Results for steel alloys, graphed in Figure 5, indicate that for a variety of steel alloys, the principal activation products (in decreasing order of importance) are Fe-55, Co-60, Ni-63, Ni-59, C-14 and Sb-125. After a significant decay period (about 30 years), the activity is dominated by Ni-63, Ni-59 and C-14. For these long-lived radionuclides, the alloys SS-304L and SS-PH are likely to yield conservative estimates for activity concentrations when the specific composition of a steel alloy is not known.

Similar assessments were also carried out for various nickel and zirconium alloys. The specific activities of steel and nickel alloys, after 30 years of decay, were found to be essentially identical and exceeded the specific activity of zirconium alloys by a factor exceeding100.

Compared with the composition of SS-304L used in this study, Evans et al [1984] developed a much more detailed composition for the alloy in order to fully assess the formation of activation products. Essentially, no difference in the estimated radionuclide activity concentrations was observed when the more detailed composition was used instead of the present composition. This demonstrates the need to be judicious in material characterisations.

It was instructive to examine the different types of activation products formed from each individual element in an alloy. Sample calculations, expressed as neutron-induced activity per kg of each element, are shown in Table 5 for elements present in SS-304 L. The results are also applicable to other metals containing these elements. The results indicate, for example, that Ni-63 is produced by the activation of cobalt, iron and nickel with the latter being the dominant source element. Table 5 can be used to assess the sensitivity of the induced radioactivity to uncertainty in source element content; for example, an increase of 1 ppm cobalt will increase the Co-60 activity by $2.2\ 10^{4}$ Ci/kg element x $10^{-6}$ kg element/kg metal alloy or 0.022 Ci per kg of the metal alloy.

## 7.0 ESTIMATES OF TOTAL ACTIVITIES FOR VARIOUS COMPONENTS

Results obtained for all components are summarised in Tables 6 and 7, based on which the following conclusions were drawn:

- The overall activity, 33 years after shutdown, was estimated to be 4.2E+16 Bq. End shields and the calandria vessel, respectively, account for approximately 60 % and 33 % of the overall activity.

- The overall activity is dominated (~95%) by Ni-63. Other radionuclides present, in order of decreasing importance, are Co-60, {Ni-59, Nb-94, C-14 and Fe-55}[1], Zr-93, Mo-93 and Cl-36. The end shields are the dominant source for C-14, Fe-55, Co-60, Ni-59, Ni-63 and Cl-36 activities.

- The pressure tubes are the principal source for Zr-93 and Nb-94. Next to pressure tubes, calandria tubes are a significant source for Zr-93. The annular shielding assembly is the principal source for Mo-93.

[1] Activities of the bracketed radionuclides are essentially similar.

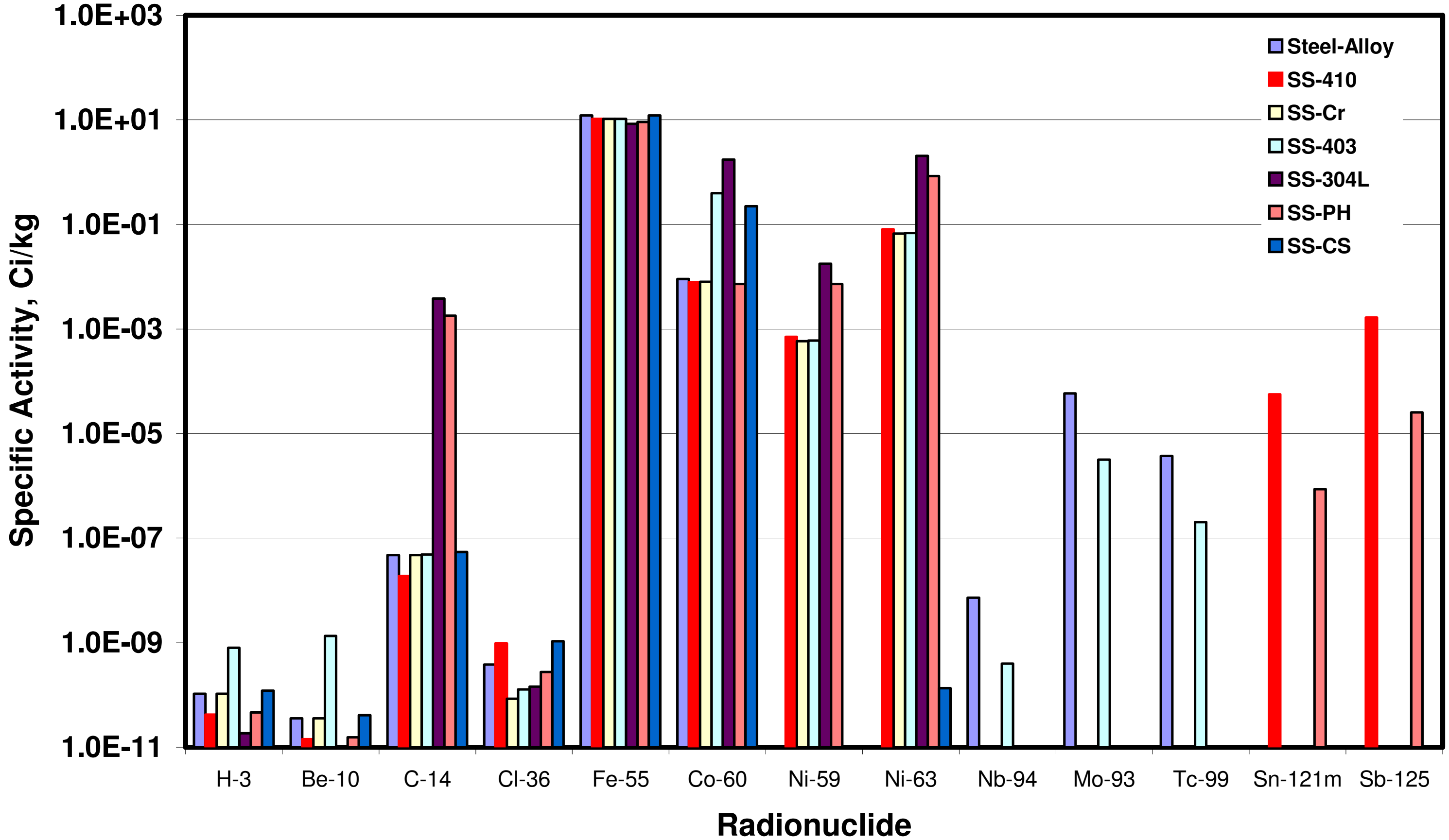



**Figure 5 Specific Activity of Activation Product Radionuclides in Various Steel Alloys**
(85% full power; irradiation time, 40 years; decay period, 2.5 years; neutron flux, 1E+12 neutrons/s/cm$^2$; thermal correction factor,1; temperature,100°C)

**Table 5 Specific Activity of Radionuclides Produced by Activation of Various Individual Elements Present in SS-304 L**

| | Half-life (Y) | Specific Activity of Radionuclides Produced by Activation (Ci per kg of Element)* | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | | **C** | **Co** | **Cr** | **Fe** | **Mn** | **N** | **Ni** | **P** | **S** | **Si** |
| Ar-39 | 2.69E+02 | | | | | | | | | 2.33E-10 | |
| Be-10 | 1.60E+06 | 1.42E-07 | | | | | | | | | |
| C-14 | 5.74E+03 | 8.40E-05 | | | | | 2.53E+01 | | | | |
| Cl-36 | 3.01E+05 | | | | | | | | | 1.99E-05 | |
| Co-58 | 1.94E-01 | | 1.71E-06 | | 1.09E-11 | | | 6.26E-04 | | | |
| Co-60 | 5.26E+00 | | 2.20E+04 | 1.28E-12 | 1.13E-01 | 4.82E-07 | | 4.84E-02 | | | |
| Cr-51 | 7.57E-02 | | | 3.44E-08 | 1.13E-12 | | | | | | |
| Fe-55 | 2.60E+00 | | | | 2.67E+01 | | | 9.38E-02 | | | |
| Fe-59 | 1.23E-01 | | 1.20E-07 | | 1.04E-06 | | | 7.76E-09 | | | |
| H-3 | 1.24E+01 | 6.42E+00 | 9.40E-11 | 7.02E-11 | 1.72E-10 | 2.52E-11 | 2.85E-06 | 2.65E-09 | 2.01E-09 | 3.58E-09 | 2.73E-10 |
| Mn-54 | 8.56E-01 | | | | 3.46E-02 | 1.47E-11 | | | | | |
| Ni-59 | 8.00E+04 | | | | | | | 3.67E-01 | | | |
| Ni-63 | 9.20E+01 | | 1.62E-04 | | 1.91E-10 | | | 4.37E+01 | | | |
| P-32 | 3.92E-02 | | | | | | | | 1.33E-09 | 4.13E-12 | 8.75E-10 |
| S-35 | 2.41E-01 | | | | | | | | | 4.83E-03 | |
| Si-32 | 6.50E+02 | | | | | | | | 1.33E-09 | 4.13E-12 | 8.75E-10 |
| Zn-65 | 6.69E-01 | | 1.46E-12 | | | | | 2.29E-06 | | | |

* Estimations were performed at a neutron flux of 5.2E+12 n/cm$^2$/s, temperature of 65°C and thermal flux weighting factor of 0. Results assume a decay period of 2.5 years after shutdown.

## Table 6 Summary of Radionuclide Inventory Data - Contribution of Key Radionuclides

| Reactor Components | Radionuclide Activity (Bq) | | | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|---|---|
| | C-14 | Fe-55 | Co-60 | Ni-59 | Ni-63 | Zr-93 | Nb-94 | Cl-36 | Mo-93 | Other | All |
| Calandria Vessel | 3.3E+13 | 2.1E+13 | 2.5E+14 | 1.3E+14 | 1.3E+16 | | | 6.4E+06 | | | 1.4E+16 |
| Calandria Tubes with Inserts | 1.1E+13 | 2.8E+12 | 6.3E+12 | 8.8E+11 | 1.6E+14 | 2.7E+12 | 2.0E+07 | 9.8E+06 | 4.4E+09 | 4.2E+12 | 1.8E+14 |
| Internal Piping | 2.9E+10 | 1.9E+10 | 2.4E+11 | 1.4E+11 | 1.2E+13 | | | | | | 1.3E+13 |
| End shields | 1.2E+14 | 4.1E+13 | 8.2E+14 | 2.0E+14 | 2.4E+16 | | | 2.2E+07 | | 1.9E+07 | 2.5E+16 |
| Shield Tank Steel Balls | 1.0E-02 | 6.8E+02 | 7.5E+02 | | | | | | | | 1.4E+03 |
| End Shield Steel Balls | 3.9E+08 | 2.4E+13 | 6.9E+13 | 7.3E+05 | | | | 5.2E+06 | | 1.4E+06 | 9.4E+13 |
| Curtain Shield Slabs | 5.2E-04 | 1.1E+01 | 4.2E+01 | | | | | | 5.0E-02 | 7.0E-03 | 5.3E+01 |
| Annular Shielding Assembly | 2.4E+08 | 5.0E+12 | 2.9E+13 | | | | 5.4E+06 | | 2.8E+10 | 6.2E+09 | 3.4E+13 |
| End Fitting Assemblies | 1.8E+08 | 9.4E+12 | 2.5E+13 | 1.6E+12 | 1.5E+14 | | 2.4E+06 | 4.5E+05 | 1.4E+10 | 1.8E+09 | 1.8E+14 |
| Closure Plugs | 1.4E+04 | 3.0E+05 | 8.1E+05 | 1.9E+05 | 1.7E+07 | 1.2E-04 | 1.4E-02 | | 7.2E+01 | 2.0E+01 | 1.8E+07 |
| Shield Plugs | 3.3E+05 | 2.4E+10 | 9.1E+10 | 3.1E+10 | 2.8E+12 | 1.1E+03 | 1.3E+05 | 6.3E+00 | 7.0E+08 | 1.5E+08 | 2.9E+12 |
| Liners & Latches | 1.3E+06 | 2.1E+11 | 4.4E+09 | 4.9E+10 | 4.4E+12 | | | | | 4.5E+09 | 4.7E+12 |
| Pressure Tubes | 2.0E+13 | 1.4E+11 | 9.2E+12 | 7.5E+10 | 1.6E+13 | 7.6E+12 | 2.9E+14 | | 1.2E+10 | 1.1E+12 | 3.4E+14 |
| Garter Springs | 6.8E+09 | 9.6E+09 | 2.9E+11 | 1.0E+12 | 2.2E+14 | 2.3E+09 | 1.6E+04 | 4.0E+04 | 1.5E+06 | 4.9E+09 | 2.2E+14 |
| Grayloc Fittings | 9.6E+00 | 6.4E+05 | 9.2E+05 | | | 1.3E-02 | 1.6E+00 | | 8.2E+03 | 1.8E+03 | 1.6E+06 |
| Bearing Sleeves | 2.0E+08 | 4.2E+12 | 1.7E+13 | | 9.8E+08 | 4.5E+04 | 4.1E+06 | | 2.4E+10 | 4.2E+09 | 2.2E+13 |
| Reactor Control Mechanisms | 2.4E+13 | 6.8E+12 | 4.9E+13 | 9.7E+12 | 2.3E+15 | 1.2E+12 | 1.0E+07 | 1.1E+07 | 8.0E+08 | 7.1E+12 | 2.4E+15 |
| **Total** | **2.1E+14** | **1.2E+14** | **1.3E+15** | **3.5E+14** | **4.0E+16** | **1.2E+13** | **2.9E+14** | **5.4E+07** | **8.5E+10** | **1.2E+13** | **4.2E+16** |

### Table 7 Summary of Radionuclide Inventory Data - Percentage Distribution of Key Radionuclides

| Reactor Components | Percentage Distribution | | | | | | | | |
|---|---|---|---|---|---|---|---|---|---|
| | C-14 | Fe-55 | Co-60 | Ni-59 | Ni-63 | Zr-93 | Nb-94 | Cl-36 | Mo-93 |
| Calandria Vessel | 1.6E+01 | 1.8E+01 | 2.0E+01 | 3.7E+01 | 3.3E+01 | | | 1.2E+01 | |
| Calandria Tubes with Inserts | 5.4E+00 | 2.4E+00 | 4.9E-01 | 2.5E-01 | 3.9E-01 | 2.3E+01 | 7.0E-06 | 1.8E+01 | 5.2E+00 |
| Internal Piping | 1.4E-02 | 1.6E-02 | 1.9E-02 | 4.0E-02 | 3.1E-02 | | | | |
| End shields | 5.8E+01 | 3.6E+01 | 6.4E+01 | 5.9E+01 | 6.0E+01 | | | 4.0E+01 | |
| Shield Tank Steel Balls | 4.9E-15 | 5.9E-10 | 5.9E-11 | | | | | | |
| End Shield Steel Balls | 1.9E-04 | 2.1E+01 | 5.4E+00 | 2.1E-07 | | | | 9.5E+00 | |
| Curtain Shield Slabs | 2.5E-16 | 9.9E-12 | 3.3E-12 | | | | | | 5.9E-11 |
| Annular Shielding Assembly | 1.1E-04 | 4.3E+00 | 2.3E+00 | | | | 1.9E-06 | | 3.3E+01 |
| End Fitting Assemblies | 8.6E-05 | 8.1E+00 | 2.0E+00 | 4.6E-01 | 3.6E-01 | | 8.4E-07 | 8.3E-01 | 1.7E+01 |
| Closure Plugs | 6.5E-09 | 2.6E-07 | 6.4E-08 | 5.5E-08 | 4.3E-08 | 1.0E-15 | 4.8E-15 | | 8.5E-08 |
| Shield Plugs | 1.6E-07 | 2.1E-02 | 7.2E-03 | 9.0E-03 | 6.9E-03 | 9.8E-09 | 4.7E-08 | 1.2E-05 | 8.3E-01 |
| Liners & Latches | 6.4E-07 | 1.8E-01 | 3.4E-04 | 1.4E-02 | 1.1E-02 | | | | |
| Pressure Tubes | 9.8E+00 | 1.2E-01 | 7.2E-01 | 2.1E-02 | 4.0E-02 | 6.6E+01 | 1.0E+02 | | 1.4E+01 |
| Garter Springs | 3.3E-03 | 8.3E-03 | 2.2E-02 | 2.9E-01 | 5.5E-01 | 1.9E-02 | 5.7E-09 | 7.4E-02 | 1.7E-03 |
| Grayloc Fittings | 4.6E-12 | 5.6E-07 | 7.3E-08 | | | 1.1E-13 | 5.4E-13 | | 9.6E-06 |
| Bearing Sleeves | 9.6E-05 | 3.6E+00 | 1.4E+00 | | 2.4E-06 | 3.8E-07 | 1.4E-06 | | 2.8E+01 |
| Reactor Control Mechanisms | 1.1E+01 | 5.9E+00 | 3.9E+00 | 2.8E+00 | 5.8E+00 | 1.0E+01 | 3.6E-06 | 1.9E+01 | 9.4E-01 |
| **Total** | **1.0E+02** | **1.0E+02** | **1.0E+02** | **1.0E+02** | **1.0E+02** | **1.0E+02** | **1.0E+02** | **1.0E+02** | **1.0E+02** |

Table 8 presents a comparison between the inventory estimates obtained in this study with the corresponding estimates[2] for each Pickering NGS-A and Bruce NGS-A [Ontario Hydro 1986a,b] reactor unit. Unfortunately, these Ontario Hydro reports were not available although a subsequent report [Dowell 1989] citing them was available. The latter presented the overall inventory of each radionuclide but did not present a breakdown between various components.

With the exception of Nb-94, estimated radionuclide activities in Darlington NGS decommissioning waste are essentially similar to those previously estimated for Pickering NGS-A and Bruce NGS-A stations. The estimated level of Nb-94 in Darlington NGS decommissioning waste appears to be a factor of 29 greater than the corresponding level at Bruce NGS-A, which in turn appears to be a factor of 19 greater than the level in Pickering NGS-A waste. Considering that pressure tubes are the principal source of Nb-94 and that each Pickering, Bruce and Darlington reactor has 390, 480 and 480 pressure tubes, respectively, with each pressure tube being of approximately similar mass, it is not evident why the Nb-94 levels at the three stations should differ so significantly.

**Table 8 Comparison Between Inventory Estimates for Key Activation Radionuclides in Pickering NGS-A, Bruce NGS-A and Darlington NGS Decommissioning Wastes**

| Radionuclide | Radionuclide Inventory in Decommissioning ILW (Bq) | | |
|---|---|---|---|
| | Pickering NGS-A [Ontario Hydro 1986a] | Bruce NGS-A [Ontario Hydro 1986b] | Darlington NGS (Present) |
| C-14 | 1.8E+14 | 1.4E+14 | 2.1E+14 |
| Fe-55 | 9.5E+13 | 1.6E+14 | 1.1E+14 |
| Ni-59 | 3.8E+14 | 3.8E+14 | 3.5E+14 |
| Co-60 | 3.0E+15 | 1.1E+15 | 1.3E+15 |
| Ni-63 | 2.8E+15 | 3.8E+16 | 4.0E+16 |
| Zr-93 | 1.6E+13 | 1.1E+13 | 1.2E+13 |
| Nb-94 | 5.3E+11 | 1.0E+13 | 2.9E+14 |

## 8.0 CONCLUSIONS

- A PC-based code called SNAP was used to estimate radionuclide activity concentrations in neutron-activated components of Darlington NGS. The code is equivalent to ORIGEN 2 but is much simpler to use.

- In general, the metal compositions for core components at Darlington fall into 3 main categories, namely, steel, nickel and zirconium alloys. For activation parameters pertinent to Darlington and a decay period of 33 years after shutdown, the total specific activities for steel and nickel alloys were observed to be essentially identical; these exceeded the total specific activity of zirconium alloys by a factor greater than 100. The activation behaviour of each metal category can be used to derive

[2] Considering 40 years of operation followed by 30 years of storage.

conservative activity estimates for various radionuclides in a component when its detailed composition cannot be easily specified.

- For each Darlington NGS unit, the overall activity, 33 years after shutdown, was estimated to be 4.2E+16 Bq. End shields and the calandria vessel account for approximately 60 % and 33 % of the overall activity, respectively.

- The overall activity of activated reactor core waste, 33 years after shutdown, was dominated (~95%) by Ni-63. Other key radionuclides are Co-60, Ni-59, Nb-94, C-14, Fe-55, Zr-93, Mo-93 and Cl-36.

- The pressure tubes and the calandria tubes are the principal sources for Zr-93. Nb-94 is associated exclusively with pressure tubes. The principal sources for Mo-93 include the annular shielding assembly, bearing sleeves, end fitting assemblies and pressure tubes

- With the exception of Nb-94, estimated radionuclide activities in Darlington NGS decommissioning waste are essentially similar with previous estimates for Pickering NGS-A and Bruce NGS-A stations. Differences in the estimated level of Nb-94 between the three stations are significant and cannot be readily explained based on the differences in reactor configuration at each station.

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