**2. Comment on the main body of REGDOC-1.2.3**
Comments on CNSC REGDOC-1.2.3, Licence Application Guide: Licence to Prepare Site for a Deep Geological Repository:

My comments are mainly based on Sections 3.1 to 3.11 of the Guide, which are referred to in the document under the heading “Waste Management,” where we read statements such as:

(i) The applicant’s management system should account for:
• data control, verification and validation
• data format
• traceability of data
• configuration control, including data, for environmental, meteorological, geological, geophysical, survey, hydrological, biological factors
• measuring and test equipment
• use and control of computer modelling
• field and laboratory work control
• calculations and analyses
• measures to ensure that the results of the site characterization are accurate, complete, reproducible, traceable and verifiable

(ii) The applicant must address:

• quantities and physical characteristics, including hazards posed to health and safety, of each substance or waste, including by-products for all substances or by-products that will be regulated or controlled, and the appropriate list of regulations governing their control

These two items are totally unacceptable because they are far too vague and therefore essentially meaningless. For example, what do the following statements really mean?

1. The applicant’s management system should account for data control, data calculations and analyses…., etc.

2. The applicant must address quantities and physical characteristics, including hazards posed to health and safety, of each substance or waste ,…., etc.

Statements such as these, using language such as “must address” or “should account for” are of no practical use in assessing, and ultimately mandating, what will be permitted for placement in the DGR, or how the waste will behave over time, and how radioactive emissions “from each substance” will be identified and characterized, both within and external to the depository.

Also of great concern is the REGDOCs use of the word “data.” The dictionary definition of this word is: Information in the form of a collection of discrete values describing specific quantities obtained by measurement, observation, or analysis. Unfortunately, REGDOC-1.2.3 provides no guidance on what “data” are required and how they should be obtained, verified or validated. And, how do we know if a “data” set is complete?

This issue has caused many significant errors in previous attempts by the Canadian nuclear industry to provide reliable estimates of radionuclide inventories in its proposed radioactive waste depositories. For example, in the period 2010 to 2014, I was able to prove that OPG and/or NWMO had seriously underestimated, sometimes by factors of more than 100, the radionuclide activities in its proposed Low and Intermediate Waste DGR, slated for construction on the Bruce Nuclear site near Kincardine in Southwest Ontario.

More recently, starting in 2017, I have discovered similar errors in the radionuclide inventory published by Canadian Nuclear Laboratories for its NSDF, proposed for construction at its Chalk River Ontario, site. In its initial 2017 EIS Report, CNL estimated there would be 996 tonnes of uranium in its NSDF, only to change this estimate in a later report to a value of about 100 tonnes; and this was done by CNL without providing a word of explanation.

Most unfortunately, I see nothing in the CNSC’s REGDOC-1.2.3 that addresses and thereby attempts to prevent a recurrence of this problem with future radionuclide inventory estimates. This is especially of concern with NWMO’s proposed spent nuclear fuel DGR. Volatile and highly mobile radionuclides such as H-3, C-14, Cl-36, etc, are notoriously difficult to measure and/or calculate, but are often presented in inventory tabulations as precise quantities that are known to within a few percent. This is entirely misleading and unacceptable. REGDOC-1.2.3. must address this issue by delineating precisely how such data should be determined and reported.

**3. Identify anticipated impacts of REGDOC-1.2.3**
REGDOC-1.2.3. must be changed to address the collection, verification and validation of radionuclide inventory data to prevent the reporting of erroneous inventories as has happened in the past.

Below is a copy of a letter I sent to NWMO concerning errors in its inventory report:

To: From:

Mr. Ken Nash Dr. Frank Greening
President and CEO 12 Uplands Avenue
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Toronto, Ontario
M4T 2S3 Tel: 905-317-4544
E-Mail greening@sympatico.ca

January 6th, 2014

Dear President Nash,

I have worked as a research scientist for the Canadian nuclear industry since 1978, first for Ontario Hydro and its successor company OPG, and subsequently for Bruce Power in Kincardine. As a radioanalytical chemist, my work has been mainly focussed on the characterization of reactor components such as pressure tubes and feeder pipes removed from Pickering and Bruce Units. Thus, over the past 35 years, I have compiled a large body of data on the radionuclide content of components that are now slated for long-term disposal as refurbishment or decommissioning waste.

I have reviewed published information pertaining to the disposal of radioactive waste from OPG and Bruce Power’s reactors, and have been especially interested in publications such as:

1. “Reference Low and Intermediate Level Waste Inventory for the Deep Geological Repository” OPG Report 00216-REP-03902-00003, issued in December 2010
2. “New Nuclear Darlington Environmental Assessment – Nuclear Waste Management Technical Support Document” OPG Report NK054-REP-07730-00027, issued August 2009
3. “Disposal Aspects of Low and Intermediate Level Decommissioning Waste” IAEA-TECDOC-1572, issued December 2007.

These reports contain tables of data on the specific activities of radionuclides in CANDU reactor refurbishment wastes. What is notable about these tables is the fact that much of the data are not based on direct measurements at all but rather on “scaling factors, used fuel ratios and neutron activation calculations”. Indeed, none of the data for pressure tubes, end fittings and calandria tubes is based on direct measurements and only a handful of radionuclides are reported to have been directly measured for steam generator waste.

It is therefore appropriate to ask a number of questions about the NWMO’s waste inventories:

(i) How were the scaling factors, used-fuel ratios and neutron activation calculations
implemented?

(ii) How do the theoretical/calculated activities compare with measured values when such
empirical data are available?

(iii) When measured data are unavailable, how are the calculated values verified?

I have attempted to answer these questions using published information on scaling factors, well-established neutron activation computational procedures and radionuclide measurements that I have made, or radionuclide data reported in the literature. I have focussed mainly on three radionuclides in pressure tube and steam generator waste – H-3, Nb-94 and Cm-244 – because these species exemplify troublesome issues I wish to address in this letter. I am providing this information to you for comment before I submit my findings to a journal for publication since I firmly believe this type of information belongs in the public domain.

I shall begin with a discussion of values of Nb-94 activities in pressure tube waste that are given in the three documents noted previously. These values, converted to units of Bq/kg are as follows:

Reference 1, generic OPG pressure tube: 7.58 × 109 Bq/kg

Reference 2, Darlington pressure tube: 1.06 × 1010 Bq/kg

Reference 3, Darlington pressure tube: 9.74 × 109 Bq/kg

Reference 3, Bruce pressure tube: 3.36 ×108 Bq/kg

Reference 3, Pickering pressure tube: 2.19 × 107 Bq/kg

Reference 3, which is based on calculations carried out by Kinectrics on behalf of the NWMO, has this comment on its own reported values:

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 differ so significantly.

What is remarkable about this statement regarding the calculated activities of Nb-94 in pressure tube waste is that no consideration is given to measured values for these activities. This is all the more puzzling because measured values of Nb-94 activities in irradiated pressure tube are readily available – See for example:

● A. W. Tarr et al. “Measurement of Carbon-14 and Other Long-Lived Radionuclides in Irradiated Zr-2.5 % Nb Pressure Tubes”. Proceedings of the Waste Management ‘94 Conference held in Tucson, Arizona, February 1994.

● E. L. Cooper et al. “Characterization of Radionuclides in Primary Heat Transport System Crud Samples and Pressure Tube Scrapes Samples from CANDU Reactors”. COG Technical Note TN-05-3053, April 2006.

Nb-94 has a 20,000 year half life and is produced in irradiated Zr-2.5% Nb pressure tubes by neutron activation of Nb-93. The Nb-94 activities in the eleven pressure tube samples measured in the previously noted references are shown in Table 1 in units of GBq/kg of Zr-Nb, ( = 1 × 109 Bq/kg). The pressure tubes in question were removed from Pickering and Bruce Units after about 10 EFPY of operation. Because of the long half life of Nb-94, (20,000 y), decay corrections are not an issue.

Table 1: Nb-94 Activities Measured in Pressure Tube Samples

Pressure Tube Sample ID
Axial Location
(meters) Nb-94
(GBq/kg ZrNb)
P3J09 2.9 7.6
P3F13 0.5 3.8
5.8 3.7
P4K10 0.5 4.1
3.2 7.7
P4N16 3.2 6.7
5.8 3.5
B5B16 1.5 7.8
4.0 9.6
B7T02 1.5 6.3
4.0 8.0

The Nb-94 activity in a pressure tube sample is related to the neutron fluence to which it has been exposed by the equation:
A94 = λ94 N°93 (σ93 / σ94) [ 1 − exp(−σ94φ t) ]
Where,

A94 is the activity of Nb-94 in Bq/g of sample
λ94 is the radioactive decay constant of Nb-94 (s−1)
N°93 is the number of stable Nb-93 atoms in the sample = (2.5/100) × (6.023 × 1023/92.9)
σ93 is the effective neutron cross section of Nb-93 = 1.66 × 10−24 cm2
σ94 is the effective neutron cross section of Nb-94 = 23.3 × 10−24 cm2
φ is the thermal neutron flux in neutrons/cm2/s
t is the irradiation time (s)

With long irradiation times, the term [1 − exp(−σ94φ t)] tends towards a value of unity and the Nb-94 activity approaches saturation activation, Asat, given by:
A94sat = λ94 N°93 (σ93 / σ94)
Using the Nb-93 and Nb-94 cross section values noted above it is readily determined that A94sat is 12.67 GBq/kg. Conversely, the neutron fluence, (φ t), delivered to a pressure tube sample may be determined from a measurement of A94.
In practice, pressure tubes never attain saturation activation of Nb-94. Thus, the Nb-94 activity of a pressure tube after a ten year exposure to a typical CANDU flux of 1 × 1014 neutrons/cm2/s is only about 50 % of its saturation value or about 6 × 109 Bq/kg. This value is consistent with the measured values given in Table 1 for samples taken near the mid-point (3 meter location) of a pressure tube.
Now some of the activities given on page 2 of this letter, (namely 7.58 × 109 Bq/kg for a generic OPG pressure tube, and between 1.06 × 1010 Bq/kg and 9.74 × 109 Bq/kg for a Darlington pressure tube), are consistent with the predicted saturation activity of 1.267 × 1010 Bq/kg. However, there is no doubt that the Nb-94 activities of 3.36 ×108 Bq/kg and 2.19 × 107 Bq/kg from page 2, which are the predicted values for Bruce and Pickering pressure tubes, respectively, cannot be correct.

This observation raises the obvious question: are other radionuclide activities reported by the NWMO also significantly in error? I intend to show that the answer to this question appears to be in the affirmative for tritium and transuranic species in a wide variety of wastes from CANDU reactors. And as with the erroneous data identified above for Nb-94, actual measurements of tritium and transuranic isotopes in samples of irradiated materials can be used to demonstrate the magnitude of the errors in the NWMO’s published data.

Tritium in Irradiated Pressure Tube Material

To date the NWMO has relied solely on calculated values to estimate inventories of individual radionuclides in pressure tube waste even though measured values of many species are readily available. What is more, as explained below, measured tritium concentrations are orders of magnitude higher than the values predicted by the first-order neutron activation calculations used by the NWMO to estimate radioactive waste inventories. The term “first-order calculations” refers to theoretical estimates involving only one tritium production pathway, namely neutron activation of deuterium impurities in pressure tubes via the D-2(n,γ)T-3 reaction. As-installed in a CANDU reactor, pressure tubes are specified to contain less than 20 mg/kg of hydrogen. It follows that the initial concentration of deuterium in a pressure tube is very low because the natural abundance of deuterium relative to hydrogen is only 0.016 %. Nevertheless, deuterium is produced by neutron activation of hydrogen at a significant rate because of the moderate, (0.33 barn), cross section of the H-1(n,γ)D-2 reaction. This leads to a slow build up of deuterium in operating pressure tubes.

A pressure tube containing an initial hydrogen concentration of 10 mg/kg hydrogen will attain a deuterium concentration of 0.625 mg/kg deuterium after 30 years of irradiation in a neutron flux of 1 × 1014 neutrons/cm2/s. This deuterium will itself undergo further irradiation to produce tritium which may be calculated to attain an activity of 5.2 ×106 Bq/kg at the end of 30 years. OPG/NWMO reports its pressure tube waste activities after a 5 year decay period to account for cooling of waste before transfer to a DGR. With 5 years decay, first-order calculations predict a tritium activity of 3.90 ×106 Bq/kg. This compares well with the OPG/NWMO reported value of 3.63 ×106 Bq/kg.

This level of agreement is encouraging; however, it becomes a moot point when we look at actual measurements of tritium in irradiated pressure tubes such as the data found in:

● A. W. Tarr et al. “Measurement of Carbon-14 and Other Long-Lived Radionuclides in Irradiated Zr-2.5 % Nb Pressure Tubes”. Proceedings of the Waste Management ‘94 Conference held in Tucson, Arizona, February 1994.

● G. R. Grant et al. “Measurement of C-14 and other Long-Lived Radionuclides in CANDU Pressure Tubes” Whiteshell Laboratory Report RC-697, November 1991

● D. L. Moir et al. “Activities of Carbon-14 and Other Long-Lived Radionuclides on the Surface and in the Metal of Irradiated Zr-2.5Nb Pressure Tubes from the Pickering Unit 4 Reactor” CANDU Owners Group Report No. COG-94-105, June 1994

Examples of measured tritium activities taken from these reports are presented in Table 2 below. For consistency with other radionuclide data in this letter, tritium activities are reported in GBq/kg, (= 1 × 109 Bq/kg).

Table 2: Tritium Activities Measured in Pressure Tube Samples

Pressure Tube Sample ID Axial Location (meters) Tritium Activity (GBq/kg ZrNb)
P3J09 2.9 2.8
P3F13 3.2 2.9
P4K10 3.2 1.7
P4N16 3.2 1.5
P4B17 3.0 1.6
P4M11 3.0 2.9
P4V13 3.0 2.2
B1L08 5.5 2.3
B1S04 5.5 2.1

The average of the tritium activities reported in Table 2 is 2.2 × 109 Bq/kg which is about 600 times higher than the OPG/NWMO reported value of 3.63 ×106 Bq/kg. The main reason for this significant under-estimation of tritium is that pressure tubes operate in high temperature and pressure D2O and pick up deuterium during operation as a result of the corrosion reaction:

Zr + 2D2O = ZrO2 + 2D2

This ingress of deuterium overwhelms the small production of deuterium from the H-1(n,γ)D-2 reaction, which as we have seen is less than 1 mg/kg after 30 years. By comparison, values of deuterium concentrations measured in irradiated pressure tubes show that up to 100 mg/kg of deuterium can build up in a pressure tube after 30 years of reactor operation due to the proposed corrosion mechanism. (See for example: B. Warr “Review and Predictions of Corrosion and Deuterium Uptake in the Body of Operating Pressure Tubes” Ontario Hydro Nuclear Report No. OH A-FC-97-113-P, July 1998).

Significantly, however, a calculation of the tritium activity expected in a pressure tube that picks up 3 mg/kg deuterium per year for 30 years results in a value of 0.8 × 109 Bq/kg, which is less than half of the observed value of 2.2 × 109 Bq/kg, suggesting that there is another, unidentified source of tritium production in irradiated pressure tubes. Lithium, which is added to a PHTS for pH control, appears to be the source of this “missing” tritium via the high cross section, (940 barns), thermal neutron reaction 6Li(n,a)3H reaction.
It is well documented that lithium isotopes are incorporated into pressure tube oxides by ion-exchange of Li+ for protons on the hydroxylated surfaces of ZrO2 crystallites:
-Zr-OH + Li+ OH- « -Zr-OLi + H2O
(See for example: B. Cox et al. “Mechanisms of LiOH Degradation and H3BO3 Repair of ZrO2 Films” Eleventh International Symposium on Zirconium in the Nuclear Industry, September 1995).
Secondary Ion Mass Spectrometry (SIMS) depth-profiling of isotopes of H, Li and Be has been carried out on the inside surface oxides of samples of irradiated pressure tube from high flux mid-channel locations. These consistently show evidence for neutron induced processes involving the transmutation sequence:
9Be ® 6Li ® 3H
Where the underlying nuclear reactions are:
b
9Be(n,a)6He ® 6Li

And,
6Li(n,a)3H

Beryllium is found at significant concentrations in the oxide scales of irradiated pressure tubes and is undoubtedly derived from the braze used to attach bearing pads to the fuel sheathing. (See for example: F.R. Greening. “The Characterization of Thick Oxide Patches on Removed Pressure Tubes: New Results for B3U11” OHT Memorandum to: P. Ellis, OHN, February 1999)
From available SIMS data it is possible to estimate the tritium activity in the B3U11 pressure tube oxide as a result of neutron activation of 6Li:
Average [6Li] » 5 x 1017 atoms/cm3 in an oxide of thickness » 35 mm
Hence, the surface concentration of 6Li in B3U11 is 1.75 x 1015 at/cm2.
25-year activation of 6Li via the 6Li(n,a)3H reaction ® 3H surface activity of 6.7 × 106 Bq /cm2
1 cm2 of oxide covers ~ 2.6 g of pressure tube so the effective tritium activity is 2.5 × 109 Bq/kg
This predicted level of tritium in the B3U11 pressure tube confirms that tritium production from 6Li significantly exceeds its production from deuterium even allowing for deuterium ingress from the zirconium corrosion reaction. Furthermore, the tritium concentration measured by high temperature oxidation of the B3U11 pressure tube sample was ~ 7 × 106 Bq /cm2, in excellent agreement with the calculated value of 6.7 × 106 Bq /cm2.
Uranium and Transuranics in Irradiated Pressure Tube Material

The activities of isotopes of uranium and transuranic elements plutonium, americium and curium in pressure tube waste are another example of data reported by OPG/NWMO that are based solely on theoretical estimates rather than actual measurements. Thus, for the data presented in Table B.3 of the report: “Reference Low and Intermediate Level Waste Inventory for the Deep Geological Repository” OPG Report 00216-REP-03902-00003, issued in December 2010, it is explicitly stated that the values for transuranic isotopes have been estimated using “used fuel ratios”. The data in question, in units of Bq per kg of irradiated pressure tube, and also converted to units of GBq per kg of initial uranium, are presented in Table 3 below. The latter unit was determined using the conversion factor 1 Bq U-238 = 80.35 µg uranium.

Table 3: OPG/NWMO Estimated Activities of Uranium
and Transuranics in Pressure Tube Waste

Radionuclide Activity
(Bq/kg ZrNb) Activity
(GBq/kg initial U)
U-235 0.363 0.000154
U-238 29.4 0.01246
Pu-238 7910 3.360
Pu-239 14,200 6.020
Pu-240 19,400 8.259
Pu-242 21.4 0.009099
Am-241 23,400 9.939
Am-242m 39.5 0.01680
Cm-244 Not Reported Not Reported

It is useful to compare the activities in column 3 of Table 3 – namely activities based on 1 kg of initial uranium – with data from calculations of the buildup of transuranic isotopes in CANDU fuel after various periods of irradiation. Such data comparisons form the basis of the “used fuel ratio” method. The report by L.J. Clegg and J.R. Coady: “Radioactive Decay Properties of CANDU Fuel Volume 1:The Natural Uranium Fuel Cycle”, Atomic Energy of Canada Report No. AECL-4436/1, January 1977, provide the required data based on ORIGEN code calculations for irradiations up to 1150 GJ/kg initial U.
ORIGEN is an isotope production and depletion code developed by Oak Ridge National Laboratories for the calculation of the concentrations of radionuclides in irradiated nuclear fuel. The two most important parameters in ORIGEN calculations are the fuel burnup – a measure of the fuel irradiation period - and the decay time. CANDU fuel is typically irradiated to a minimum burnup of 7500 MWd/Mg initial U, (650 GJ/kg U), equivalent to an irradiation period of about 200 days. However, ORIGEN data in AECL’s 4436/1 report extend to fuel burnups of 13,300, MWd/Mg initial U, equivalent to 350 days of irradiation.
In order to make predictions of the abundance of transuranic isotopes in irradiated pressure tubes, ORIGEN data for CANDU fuel irradiated up to 350 days and decayed 5 years have been compiled from AECL’s 4436/1 Report and are shown in Table 4 below.

Table 4: ORIGEN Predictions for CANDU Fuel
Isotopic Species Specific Activity for Specified Exposure Time
(GBq/kg initial U)
40 days 150 days 200 days 240 days 350 days
Pu-238 0.081 1.42 2.63 4.29 9.11
Pu-239 2.74 5.39 5.92 6.16 6.34
Pu-240 0.89 5.90 8.18 10.33 14.17
Am-241 0.270 3.44 5.21 7.06 10.66
Cm-244 - 0.13 0.381 1.04 4.84

The most important transuranic isotopes (with half-lives greater than one year) are as follows:
Isotope Half-Life (yr)
Pu-238 87.7
Pu-239 24,100
Pu-240 6,560
Pu-241 14.4
Am-241 433
Cm-244 18.1

For the purposes of comparisons between calculated and measured data it is preferable to look at data for long-lived isotopes such as Pu-239 or Pu-240. The process by which transuranic isotopes are produced in a CANDU reactor is neutron absorption by the natural uranium fuel which contains 99.27 % U-238. This leads, through β-decay of short-lived Np-239, to the formation of the important long-lived α-emitting isotope Pu-239:

U-238 (n,γ) U-239 → Np-239 → Pu-239

Further irradiation of Pu-239 results in the slow build up of additional long-lived α-emitting isotopes such as Pu-240, Am-241, and Cm-244. The time-dependent variation in the concentration of Pu-239 is given by the equation:

dN239/dt = No238 σ238 φ - N239 σ239 φ
Where:
N239 is the number of Pu-239 atoms per unit mass of fuel
No238 is the initial number of U-238 atoms per unit mass of fuel
σ238 is the total neutron capture cross section of U-238
σ239 is the total neutron capture cross section of Pu-239
φ is the thermal neutron flux
After a prolonged in-reactor exposure of uranium, the buildup of Pu-239 saturates and eventually reaches an equilibrium where dN239/dt = 0. The equation above then reduces to:
N239 / No238 = σ238 / σ239 = 0.0027
Thus the equilibrium concentration of Pu-239 in CANDU fuel is 2.7 g per kg of initial U-238. This concentration may be converted to a Pu-239 saturation activity, Asat(Pu-239) as follows:
Asat(Pu-239)/kg U = (2.7 × 6.02 × 1023 × 0.693) / (239 × 2.41 × 3.156 × 1011)
Asat(Pu-239)/kg U = 6.2 × 109 Bq/kg U
This value for Pu-239 is very close to the OPG/NWMO value of 6.02 × 109 Bq/kg U reported in Table 3 above. Converting this activity to a per kg of Zr basis, the OPG/NWMO values for U-238 and Pu-239 in pressure tube waste are consistent with an ORIGEN code calculation for a 350-day irradiation of a Zr-2.5%Nb pressure tube containing 2.4 mg/kg initial uranium. This concentration is well above the specification of 0.3 mg/kg for uranium impurity in Zr-2.5%Nb pressure tube material and suggests that in-reactor contamination of pressure tube surfaces by “tramp” uranium is responsible. But are there experimental data on the concentration of uranium and transuranic species in irradiated pressure tube samples to support this hypothesis? There certainly are suitable data but as demonstrated below, measured values of uranium and transuranics in pressure tubes are consistently much higher than the OPG/NWMO estimates.

Uranium and transuranics in pressure tubes are determined by techniques such as mass spectrometry and alpha spectrometry. Alpha spectra are acquired with an ion-implanted silicon detector typically having an energy resolution ~ 0.02 MeV (FWHM). In view of this practical limitation, it is not possible to resolve certain isotope pairs in an alpha spectrum – most notably Pu-239/Pu-240 and Cm-243/Cm-244. For this reason it is standard practice to report Pu-239 and Pu-240 as their combined activity. In the case of Cm-243 and Cm-244, ORIGEN code calculations show that the Cm-243 activity is typically less than 2 % of the corresponding Cm-244 activity which is therefore reported as Cm-244 alone without introducing significant error.
One of the most useful sets of data on the concentration of uranium and transuranic species in irradiated pressure tube samples is in the report by E. L. Cooper et al. “Characterization of Radionuclides in Primary Heat Transport System Crud Samples and Pressure Tube Scrapes Samples from CANDU Reactors”. COG Technical Note TN-05-3053, April 2006. Data from this report are presented in Table 5.
Table 5: Uranium and Transuranic Radionuclides in Bruce Pressure Tube Samples

Sample ID U-238
(mg/kg ZrNb) Pu-238
(Bq/kg ZrNb) Pu-239/240
(Bq/kg ZrNb) Am-241
(Bq/kg ZrNb) Cm-244
(Bq/kg ZrNb)
B5B16 (1.5 m) 25 280,000 190,000 470,000 73,000,000
B5B16 (4.0 m) 12 160,000 130,000 470,000 74,000,000
B7T02 (1.5 m) 20 250,000 170,000 190,000 22,000,000
B7T02 (4.0 m) 12 130,000 100,000 140,000 30,000,000
Data on uranium and transuranics on the surface of CANDU pressure tubes have also been obtained from SIMS (Secondary Ion Mass Spectrometry) analysis of a Bruce pressure tube inside surface oxide, (Data reported here are for a B3U11 pressure tube sample analyzed in 1999). The relative peak intensities in the SIMS spectra have been normalized to unity for U-238, converted to activities using the relation A = Nλ, and multiplied by a factor of 2.35 µg for direct comparison with OPG/NWMO estimates. The results are collected in Table 6 below together with averaged values of the experimental data reported in Table 5.

Table 6: A Comparison of OPG/NWMO Data with SIMS and
Alpha Spectrometric Measurements on Bruce Pressure Tubes

Radionuclide Estimated Values Measured Values
OPG/NWMO Activity
(Bq/kg ZrNb) Averaged Data from Table 5 (Bq/kg ZrNb) SIMS Data for B3U11
(Bq/kg ZrNb)
U-235 0.363 - 0.701
U-238 29.4 214 29.4
Pu-238 7910 205,000 -
Pu-239 14,200 62,000 104,000
Pu-240 19,400 85,000 133,000
Am-241 23,400 318,000 146,000
Cm-244 Not Reported 50,000,000 14,900,000

A number of important conclusions may be drawn from the data presented in Table 6:
● The measured activities of alpha-emitting transuranic radionucldes in irradiated pressure tubes are considerably higher than the OPG/NWMO “used fuel ratio” estimates
● Cm-244, which is not included in the OPG/NWMO’s estimates, is by far the highest alpha activity measured in irradiated pressure tube samples
● There is a high degree of variability, (by up to a factor of 3), between measurements from different pressure tube samples
These observations are further supported by data for Pickering pressure tube samples taken from the report by J.D. Chen et al. “Measurement of C-14 and other Long-Lived Radionuclides in Irradiated Zr-2.5Nb Pressure Tubes from Pickering Units 3 and 4” CANDU Owners Group Report No. COG-93-22, June 1993, as summarized in Table 7 for samples taken near the 3 meter axial location.

Table 7: Transuranic Radionuclides Measured in Pickering Pressure Tube Samples

Radionuclide Sample Designation
P3J09
(Bq/kg ZrNb) P3F13
(Bq/kg ZrNb) P4K10
(Bq/kg ZrNb) P4N16
(Bq/kg ZrNb)
Pu-238 47,000 32,000 44,000 77,000
Pu-239 + Pu-240 54,000 54,000 71,000 110,000
Am-241 < 10,000 200,000 200,000 200,000
Cm-244 18,000,000 2,400,000 13,000,000 5,500,000

A comparison of the pressure tube data in Tables 6 and 7 shows that the alpha-activities in Bruce samples are somewhat higher than the activities of the equivalent species in Pickering samples. Nevertheless, these measured values are consistently higher than the OPG/NWMO estimated values by a factor of at least 3. More importantly, however, the reported OPG/NWMO values imply that Cm-244 is entirely absent from irradiated pressure tubes when in fact this radionuclide accounts for more than 90% of the alpha activity in this type of waste. It is worth noting that Cm-244 is an important component of radioactive waste because it decays to another alpha-emitter, Pu-240, and is a significant neutron source through spontaneous fission and the associated (α,n) reactions with O-17 and O-18 in metal oxides in the waste matrix.

Finally there are additional reasons to reject the entire set of OPG/NWMO data for pressure tube waste as tabulated in “Reference Low and Intermediate Level Waste Inventory for the Deep Geological Repository”, (OPG Report 00216-REP-03902-00003, issued in December 2010), because many of the measured fission product and activated corrosion product activities are orders of magnitude higher than estimated values. This is illustrated in Table 8 for a selection of published data on gamma-emitting radionuclides in removed pressure tubes.

Table 8: Comparison of Measured and OPG/NWMO Estimated Values of Gamma-
Emitting Radionuclide Activities in Pressure Tubes

Type of Data Sample ID Radionuclide Activity (MBq/kg ZrNb)
Mn-54 Co-60 Cs-134 Cs-137 Sb-125
Estimated OPG/NWMO 4.94 2540 1.81 0.025 36.3
Measured B5B16 (1.5 m) 780 5300 190 170 -
Measured B5B16 (4.0 m) 730 6800 200 160 -
Measured B7T02 (1.5 m) 690 5800 100 62 -
Measured B7T02 (4.0 m) 750 6800 90 70 -
Measured P3J09 (2.9 m) - 2800 - 50 -
Measured P3F13 (0.5 m) - 1500 - 35 61
Measured P3F13 (3.2 m) - 3100 - 340 190
Measured P3F13 (5.8 m) - 1700 - 19 110
Measured P4K10 (0.5 m) - 2800 - 12 -
Measured P4K10 (3.2 m) - 5000 - 130 -
Measured P4N16 (3.2 m) - 2500 - 110 -
Measured P4N16 (5.8 m) - 1300 - 20 -
Measured P4B17 (3.0 m) 71 3500 - 47 -
Measured P4M11 (3.0 m) 110 5600 15 50 200
Measured P4V13 (3.0 m) 120 5200 23 43 88

The data in Table 8 clearly demonstrate that OPG/NWMO’s gamma-emitting fission and activated corrosion product estimates for pressure tube waste are consistently less than the measured values, sometimes by a factor of more than 100.

The Overall Accuracy of OPG/NWMO`s Inventory Estimates

On April 18th, 2012 a meeting was held between the CNSC, OPG and the NWMO at which the CNSC raised its concern that the uncertainties and necessary conservatism in radionuclide inventory estimates for low and intermediate level waste have not been adequately addressed by OPG and the NWMO. These concerns are described in detail in two Environmental Impact Statement Information Requests - EIS-01-06 and EIS-01-20 – submitted by the Deep Geologic Repository Joint Review Panel of CEAA.

OPG/NWMO first addressed these questions in: “Reference Low and Intermediate Level Waste Inventory for the Deep Geological Repository” OPG Report 00216-REP-03902-00003, issued in December 2010, where we find the following statement:

“ORIGEN calculates radionuclides to within a factor of 3 in general (some exceptions), and Zr and Nb within 30% for a specific irradiation history. There is also some indication that the ORIGEN results are generally higher than actual inventories by a factor of 1 to 3”

Unfortunately, no evidence is provided for the claim that neutron activation calculations using the ORIGEN code tend to give high numbers. Nevertheless, OPG/NWMO provide an overhead for the April 18th, 2012 meeting which is reproduced below as Table 9.

Table 9: OPG Overhead

“Data on ORIGEN Accuracy for Relevant Nuclides/Materials”

Nuclide Accuracy (Calculated/Measured) Source Reference
Mn-54, Zr-95, Nb-95,Sn-113, Sb-125, Hf-181 Within factor of 1.5 Pickering
Unit 2
Pressure Tube Aydogdu et al.
(1989)
Co-60 and Sb-125 Within factor of 3
Ta-182 Within factor of 10
Nb-94 2-3 Pickering Unit 3 & 4 Pressure Tubes Chen et al (1993)
Moir et al (1994)
Fission Products ~ 10 % PWR/BWR fuel SKB (2010)

There are some obvious concerns with these uncertainty estimates from OPG. For example:

● Data for Zircaloy-2, (used in pressure tubes for Pickering Units 1 & 2) are mixed with data for Zr-2.5%Nb. These alloys have quite different levels of trace elements such as Ni, Cr, Sn and Ta

● Data for PWR/BWR fuel, (which is enriched in U-235), are used instead of data for non-enriched CANDU fuel

However, there are other uncertainties associated with the application of neutron activation calculations to irradiated pressure tubes that need to be considered including:

● Uncertainties in the application of decay corrections to measured and calculated data

● Measured data are only available for tubes with irradiations up to about 15 years

● Variability in the concentration of surface contaminants such as uranium and transuranics

Nevertheless, even with all these uncertainties taken into account, neutron activation calculations should (in principle!), be capable of predicting radionuclide inventories in reactor waste to within a factor of about 10. Clearly, as shown in this letter, OPG/NWMO’s estimated inventories do not meet this expectation, let alone the more ambitious claims of accuracies to within a factor of 3 for the radionuclides listed in Table 9. Remarkably, however, OPG’s inability to reliably estimate radionuclide inventories is not due to any of the sources of uncertainty noted above, but rather to a failure to recognize how species in primary heat transport D2O, such as Li, Be, Fe, Co and U, interact with and become attached to pressure tube surfaces.

Significantly, OPG do claim to address the issue of surface contamination of pressure tubes - See for example page 29 of their report: “Reference Low and Intermediate Level Waste Inventory for the Deep Geological Repository” OPG Report 00216-REP-03902-00003, issued in December 2010, where we read:

“The contribution from surface contamination is included for pressure tubes
based on data from outlet feeder pipes”

However, this is simply not a valid approach because feeder pipes are made of carbon steel and form a magnetite surface oxide layer as opposed to Zr-2.5% Nb pressure tubes which form a zirconium oxide layer on their inside surface during reactor operation, and there is no reason to expect these oxides to exhibit the same ion exchange properties for dissolved species in a PHTS.
Furthermore, the pickup of Co-60 by pressure tubes has been measured directly as described by F. R. Greening in “Post Irradiation Investigation of Corrosion and Deuterium Pickup by Zr-2.5% Nb Alloy Pressure Tubes: Isotope Tracers in Inside Surface Oxides” CANDU Owners Group Report No. COG-96-618, Dec 1996. This report shows that Co-60 accumulates in irradiated pressure tubes by two mechanisms:

(i) Neutron activation of cobalt present as a trace impurity in the base Zr/Nb alloy.
(ii) Pickup of Co-60 by its incorporation into pressure tube inside surface oxides

The relative contributions to the Co-60 activities observed in irradiated pressure tubes have been evaluated by radiochemical analyses of Co-60 in tube sections cut to exclude the inside surface oxide. For pressure tube P3M11, a base metal Co-60 activity of 36 µCi/g was measured compared to 60 – 80 µCi/g for samples with the ID oxide still present. This shows that about 50 % of the Co-60 activity in a pressure tube is located in the surface oxide which, in the case of the P3M11 tube, is only 10 µm thick. This implies there is at least a 100-fold enrichment in the Co-60 concentration in the surface oxide compared to the base metal and explains why OPG’s first-order neutron activation calculations of Co-60 activities in irradiated pressure tubes underestimate the true activity of this species.

A similar situation prevails for uranium fission products and transuranics in irradiated pressure tubes because production of these species by activation of uranium impurity in the base metal is much less than the production from uranium incorporated into a pressure tube’s inside surface oxide during reactor operation. And the behavior of uranium on the surface of a pressure tube is quite different to its behavior on a feeder pipe surface, not only because the oxide substrates are chemically quite different (ZrO2 vs.Fe3O4), but also because uranium undergoes continuous neutron irradiation in the former case, but not in the latter.

Conclusions

The specific activities published by OPG and the NWMO for radionuclide inventories associated with CANDU pressure tube refurbishment waste are seriously underestimated, sometimes by factors of more than 100. This discrepancy stems from undue reliance on calculations that do not properly account for neutron activation processes occurring in pressure tube material exposed to high neutron fluences in chemically aggressive environments such as high temperature lithiated D2O. This problem could have been avoided by the authors of the calculations had they simply compared the results of their calculations with readily available data from direct measurements of pressure tube sections. Nevertheless, direct measurement of radionuclides in other types of CANDU waste, (e.g. steam generators), reveals there may also be problems with empirical data due to the variability seen in many directly measured radionuclide inventories. For example, even the relatively well-characterized radionuclide Co-60 shows a variability of a factor of 20 between different steam generator samples. It then becomes very problematic to use Co-60 data to develop scaling factors for other, difficult-to-measure radionuclides. I therefore urge you to address these issues and amend your radionuclide waste inventories accordingly.

Sincerely,

Dr. F. R. Greening

c.c. Dr. M. Binder, Canadian Nuclear Safety Commission
Dr. S. Swanson, Canadian Environmental Assessment Agency