

IN-SITU GAMMA SPECTROMETRY OF SPENT RESIN AND FILTER ELEMENTS AT CANDU[®] STATIONS

Y. Verzilov¹ and A. Husain¹

¹Kinectrics Inc., Toronto, Ontario, Canada

yury.verzilov@kinectrics.com, aamir.husain@kinectrics.com

Abstract

Kinectrics routinely performs gamma spectrometry surveys of key reactor system components during outages at CANDU[®] stations to monitor build-up of radiation fields. Recently, the scope of the surveys was expanded to include monitoring of spent resins and filters. Characterization data for these media are also of interest for the safety case and the licensing of the Deep Geological Repository which is being developed by OPG to dispose its Low and Intermediate Level wastes.

This paper presents a description of the new in-situ gamma spectrometry technique. Pros and cons of the new approach and the methodologies employed previously are discussed. The new technique yields only gamma activity data, which must be pooled together with alpha and beta activity data obtained from characterization of collected samples. Nevertheless, its simplicity permits ease of data collection with minimal impact on station resources and with negligible dose expenditure.

New results obtained are compared with previous data.

1.0 Introduction and background

Figure 1 shows a simplified schematic of the two-loop CANDU[®] Primary Heat Transport (PHT) System. The two figure-of-eight loops provide bi-directional flow through the reactor core. During operation, corrosion and wear products from various PHT system surfaces are transported by the coolant into the reactor core, where they undergo activation. These, along with fission products and actinides released from defective/tramp fuel, deposit on out-core reactor surfaces resulting in the development of gamma radiation fields. Mechanical filters and ion exchange columns located in a side stream purification flow circuit (not shown in Figure 1) help to control radionuclide deposition on PHT system surfaces and hence radiation field growth on system components.

For the purpose of monitoring radiation fields, Kinectrics routinely conducts Outage Activity Transport Monitoring surveys of key reactor system components such as the reactor face, feeder cabinets and steam generators at various stations. A research program for improved characterization of reactor radiation fields, sponsored by the CANDU Owners Group (COG), has also been underway at Kinectrics since about 2007 [1, 2].

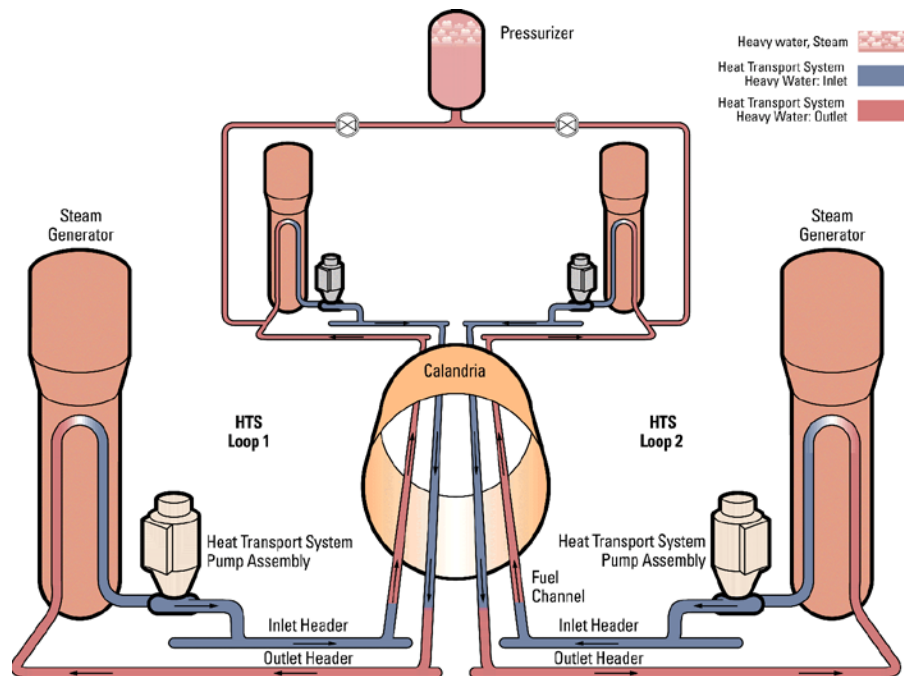


Figure 1. Simplified Schematic of the Two Loop Primary Heat Transport System

As part of the COG work program, gamma activity measurements of spent filters and spent ion exchange resins from the purification system were recently initiated. The methodology employed and the results obtained to date are described in this paper. Results for spent resins are compared with previous measurements made by directly sampling spent resins from station storage tanks [3, 4], while results for spent filters are compared with gamma activity measurements performed at the Western Waste Management Facility(WWMF) during the unloading of spent PHT filters into an underground storage pipe liner [5].

The new approach developed for measuring spent filter and spent resin activities is also of significant interest to the Nuclear Waste Management Organization (NWMO), which has responsibility to develop the planned Deep Geological Repository (DGR) for Low and Intermediate Level wastes on behalf of Ontario Power Generation (OPG). Kinectrics has been involved in waste characterization activities in support of the DGR on an ongoing basis since about 1999.

Data presented in this paper originate from a number of OPG stations. For proprietary reasons, they are presented without disclosing their specific source, although the sources are differentiated as Station-A to Station-E.

2.0 PHT purification system resins at OPG stations

Table 1 presents a summary of the PHT purification system configuration at various OPG stations.

Table 1: Purification System Configuration at Various OPG Stations

Station	No. of Banks	System Configuration	
		Mechanical Filter	Ion Exchange Resin Columns*
A	1	2 filters 13 elements per filter Pore size (Filter 1): 0.1 µm Pore size (Filter 2): 0.45 µm	3 columns each containing 4 ft ³ NRW-37LiLC mixed bed resin & 6 ft ³ NRW-600LC anion resin
B	2	1 filter per bank 13 elements per filter Pore size (Bank 1): 0.1 µm Pore size (Bank 2): 0.45 µm	3 columns each containing 3 ft ³ NRW-37LiLC mixed bed resin & 4 ft ³ NRW-600LC anion resin
C	2	2 filters per bank 21 elements per filter Pore size (Filter 1): 0.1 µm Pore size (Filter 2): 1 µm	3 columns per bank each containing 1 ft ³ NRW-37LiLC mixed bed resin & 6 ft ³ NRW-600LC anion resin
D	2	2 filters per bank 21 elements per filter Pore size (Filter 1): 0.1 µm Pore size (Filter 2): 2 µm	3 columns per bank each containing 1 ft ³ NRW-37LiLC mixed bed resin & 6 ft ³ NRW-600LC anion resin
E	2	1 filter per bank 13 elements per filter Pore size : 0.1 µm	3 disposable 4.5 ft ³ columns per bank

*NRW-37LiLC: Purolite lithiated mixed bed resin for pH control;
NRW-600 LC: Purolite OH⁻ form anion resin

The PHT purification systems at OPG stations generally utilize bead type ion exchange resins. Strong acid cation (SAC)/ strong base anion (SBA) mixed bed resins in the Li⁺/OH⁻ form (Purolite NRW-37LiLC) is typically used in conjunction with strong base (OH⁻ form) anion resins. The resins are physically layered so that NRW-37LiLC is at the bottom of the column while NRW-600LC is at the top; at Station-C and Station-D, the layers may be configured in the reverse order.

While Station-E PHT purification system uses disposable ion exchange columns, the other stations use permanent resin service columns. New resin is slurry loaded into these columns; when spent, the resins are slurried out from the service column into the de-deuteration hopper, where the heavy water associated with the resin is displaced by light water and recovered. The de-deuterated resins are then transferred from the hopper to the station storage tank.

Submicron sized filters are commonly used in PHT purification systems at OPG stations. They employ Pall's pleated glass fiber membrane filters of different pore sizes. Over the years, a transition to filters with tighter pore size has occurred. Currently, the smallest pore size filter in use is Pall's Ultipor 0.1 µm filter. In general, filter cartridges are replaced based on a differential pressure criterion. The spent filter cartridges are transferred from the service vessel in to an in-station flask and subsequently into the transportation package for shipment to the WWMF.

PHT purification system operating practices differ from station to station. At Station-A, for instance, only one filter (0.1 μm) and both ion-exchange columns are required during normal steady-state operation; at other times (e.g. during PHT system warmup, reactor power ramp up, shutdown or cool down), when crud bursts may occur, the second filter (0.45 μm) is also put into service. Occasionally, because of high differential pressure across the filter, the flow may be routed directly to the ion exchange columns, bypassing the filter.

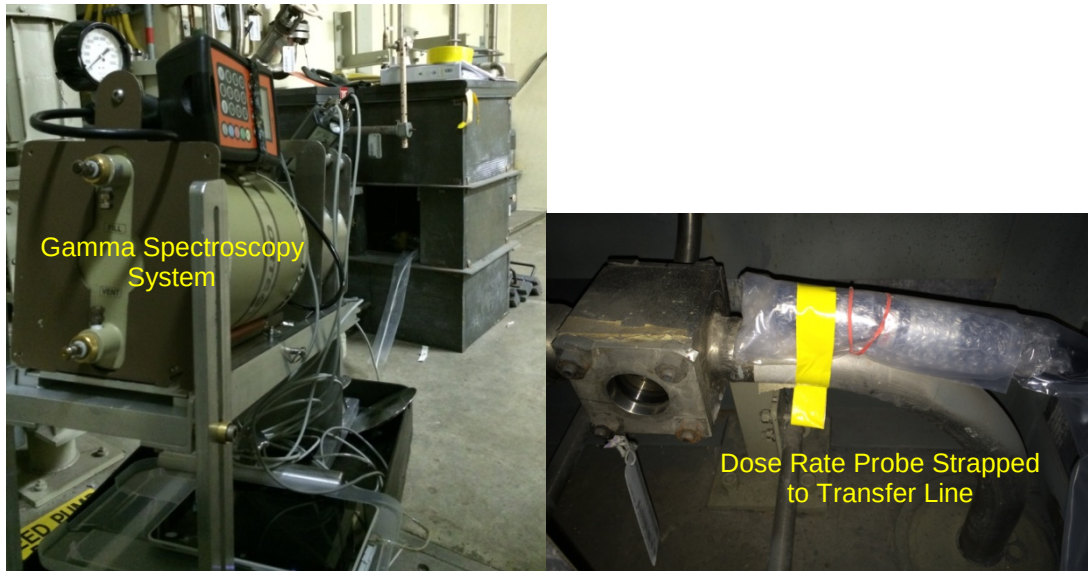
3.0 Approach for measurement of spent resin and filter activities

3.1 Spent resin activity

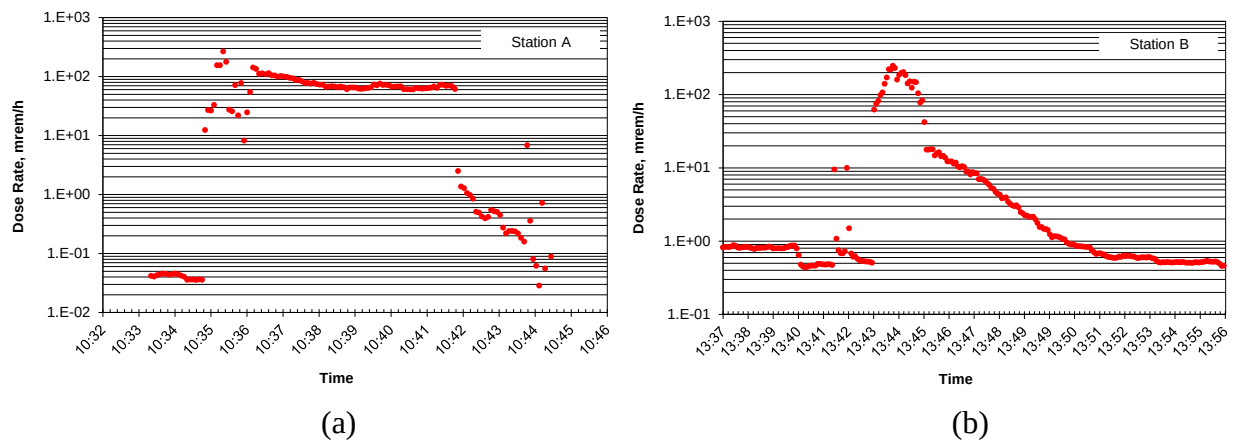
In principle, ion exchange resin activity may be ascertained by 1) monitoring the activity difference between the column influent and effluent streams, or 2) by directly measuring the resin column activity. Both approaches were assessed and shown not to be feasible, in the first instance, because of the relatively high background field and, in the second instance, because the columns are heavily shielded. On the other hand, monitoring the activity of freshly discharged de-deuterated resins during slurry transfer from the hopper to the spent resin storage tank was considered to be feasible.

Accordingly, measurements were performed using a mobile gamma spectroscopy system consisting of a 10 % Hyper Pure germanium detector, a liquid nitrogen dewar and a digital multichannel analyser (digiDART). Figure 2(a) shows a view of the room at Station-A where gamma spectroscopy measurements were performed on the resin slurry transfer line; the close up view (see Figure 2b) also shows a survey probe mounted on the transfer line for on-line monitoring of dose rates during the transfer.

Figure 3 (a) shows the contact dose rate variation recorded during resin transfer at Station-A; the dose rate increased rapidly from background levels to about 100 mrem/h and remained at this level over a significant portion of the overall transfer duration (approximately 10 min). The plateau region observed in Figure 3(a) was, however, characteristically absent in the dose rate trend observed at Station-B (see Figure 3b). Finally, the trends observed at Station-C were characterized by dual peaks. These trends possibly result from a combination of factors such as 1) the degree of intermixing, between the dual layers of resin originally present in the service, experienced during the initial transfer of resin from the service column to the de-deuteration hopper and its subsequent transfer to the storage tank, 2) the specific radionuclides bound to each type of resin and 3) the variable resin/water slurry composition.



(a) (b)
Figure 2: Gamma Spectroscopy Setup for Measuring Spent Resin Activity



(a) (b)
Figure 3: Measured Contact Dose Rates Recorded on Transfer Line during Spent Resin Transfer at a) Station-A and b) Station-B

Gamma spectra recorded during the resin transfers were interpreted as follows to obtain the average radionuclide activity concentrations for the overall spent resin column:

- The recorded spectra peak count rates (cps) were normalized into equivalent volumetric units (cps/cm³) based on the volume of resin contained in the service column (10 ft³ Station-A resin, otherwise 7 ft³ resin).

- A value for the conversion factor (Bq/cm^3) per (cps/cm^3) was obtained using MERCURAD [6] considering the applicable pipe geometry (diameter 2.5 inch, pipe length in field of view of ~ 1 m and pipe-detector distance of ~ 0.5 m) and a resin/water ratio of 0.5 (because water and resin have similar densities, sensitivity of the conversion factor estimate to the selected resin/water ratio was low).
- The radionuclide activity (Bq/cm^3) obtained by applying the conversion factor to the measured data was decay corrected to the column's end-of-service date.

3.2 Filter activity

The only feasible option for measuring filter activity is when the spent filter cartridge is exposed (outside its housing) during its post-service handling. Typically, the filter cartridge is removed from the top of the service column and placed in a bagged over-pack (drip can), the latter being then top-loaded into the in-station flask; this transfer occurs in the purification room. Thereafter, the in-station flask is moved into the transfer room, where the over-pack is recovered and top loaded into the transportation package.

Figure 4 shows a view of the transfer room at Station-B. With the gamma spectroscopy system pre-staged in the transfer room, a spectrum of the suspended filter over-pack can be conveniently obtained (count duration about 15 min) from a distance of about 3 m.

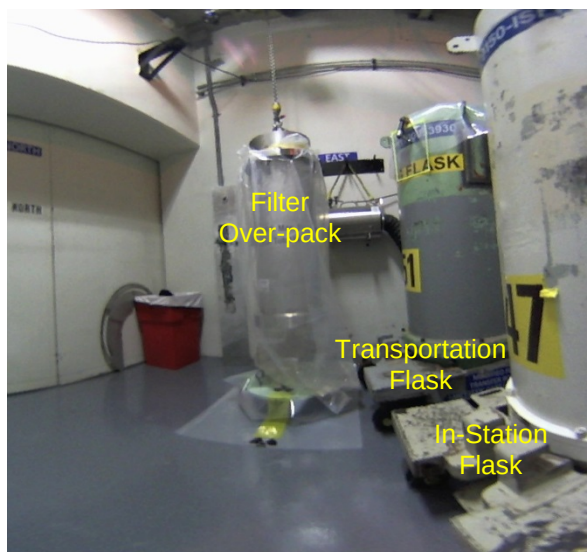


Figure 4: Spent Filter Handling in the Transfer Room at Station-B

The gamma spectroscopy data were interpreted using MERCURAD [6]. This yielded a relationship between counts/s and activity in Bq/cm³, thus allowing the measured count rate to be converted in an equivalent volumetric activity; the total filter activity was then obtained using the volume of the filter.

4.0 Measured resin activity results

As per Table 1, transfers at Station-A involve the slurring of 10 ft³ resin while transfers at other stations involve 7 ft³ of resin. Table 2 presents particulars of the resin transfer measurements undertaken at three OPG stations in 2014-15. Note the following:

- The service duration of the various PHT resin columns varied significantly (250 - 800 d).
- As shown in Column 3, the spent resin in some cases was slurried out soon after being spent, while in the other cases, over 400 d elapsed.
- With few exceptions, the observed dose rates were highly variable (based on large ratio between the maximum (Column 4) and the average (Column 5) observed dose rates).
- The Station-C contact dose rates were uncharacteristically low compared to those observed at Station-A and Station-B. This may largely be due to the relatively high anion resin component and partly due to the extended elapsed duration between the resin end-of-service and transfer dates.
- In general, the transfer durations were variable and did not correlate with the volume of resins transferred, implying variable water/resin ratios during the slurry transfers.

Table 2: Particulars of Spent Ion Exchange Resin Transfers at OPG Stations

Station	Service Duration (d)	Duration Between Transfer Date and End of Service Date (d)	Contact Dose Rate on Transfer Line (mrem/h)		Time for Resin Transfer from Hopper to Storage Tank
			Maximum	Average	
A	725	268	266	79	6 min 36 s
	453	84	960	199	5 min 35 s
	520	12	439	136	2 min 05 s
	305	22	44	31	5 min 26 s
	677	70	382	130	3 min 15 s
B	797	6	247	88	3 min 35 s
	486	166	100	11	6 min 50 s
	566	144	116	33	3 min 15 s
C	251	414	3.8	2.3	7 min 30 s
	251	415	3.9	1.9	6 min 20 s

Average gamma spectrometry results obtained for the three stations are compared in Figure 4. The activity of Station-A and Station-B resins were generally comparable while those for Station-C were significantly lower. Note that as per Table 1, the ratio of mixed bed to anion resins is about 4/6 for Station-A and Station-B resins, and 1/6 for Station-C resins. Thus, for similar coolant radionuclide concentrations, one would expect the Station-C resins to have lower concentrations of cationic species such as Co-60, but higher concentrations of anionic species such as Sb-124 & Sb-125. Overall, for each station, Co-60 represented a relatively small percentage of the resin activity, with Cs-137 being generally the dominant longer-lived radionuclide present.

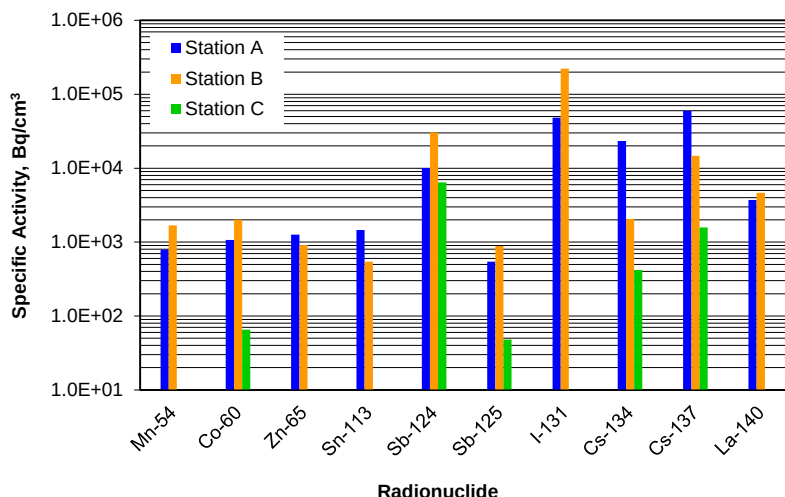


Figure 4: Comparison between Measured Spent Resin Activities for Various Stations

Table 3 presents a comparison between the present mean activity results for Station-A and mean activity results for resin samples extracted from Station-A PHT resin storage tanks [6]. The ratio between the two sets indicates that, in general, the two approaches yielded consistent results; the more significant discrepancies are observed in the cases of Mn-54 and Sb-124. While the present data were decay-corrected to the end-of-service date for each column, the measured sample results (except Sn-113 and Sb-124) were decay-corrected to the start of the resin storage date. The difference between these two dates may be significant as shown in Table 2, although this would not explain the discrepancy in the Mn-54 results.

Table 3: Comparison between Sample and In-Situ Measurement Results

Nuclide	Sample	In-Situ (Bq/cm ³)	Ratio A/B
Mn-54	8.2E+03	7.1E+02	11.6
Co-60	1.9E+03	5.8E+02	3.3
Zn-65	4.2E+03	1.1E+03	3.8
Sn-113*	5.0E+02	1.7E+03	1/3.4
Sb-124*	1.1E+03	9.3E+03	1/8.5
Sb-125	4.4E+02	7.8E+02	1/1.8
Cs-134	1.3E+04	1.5E+04	1/1.1
Cs-137	3.3E+04	3.7E+04	1/1.1

*Data for short-lived radionuclides (Sn-113 and Sb-124) were not corrected for storage duration in tank.

5.0 Measured filter activity

Table 4 presents particulars of two filter measurements undertaken at Station-E in 2015-6. The Station-E filter basket assembly, supplied by Pall, has 14 glass fibre elements.

Table 4: Particulars of Filter Measurements undertaken at Station-E

Filter	In-service Date	End-of-Service Date	Duration in Service (d)	Measurement Date
1 (0.1 µm)	14-May-2013	06-Jun-2015	753	02-Nov-2015
2 (0.45 µm)	03-Dec-2013	08-Jan-2016	650	04-May-2016

Measurement results, decay-corrected to the end-of-service date are presented in Table 5 along with a comparison with previously measured data[5] for a Station-APHT filter. Note the following:

- Co-60, Zr/Nb-95 and Fe-59 were the dominant radionuclides present on the filter.
- The Nb-95/Zr-95 ratio for the Unit 1 and Unit 4 filters were 2.3 and 2.0, respectively. At equilibrium, the ratio is expected to have a value of 2.2. As previously shown [2], if the radionuclides originate from pressure tubes, then the ratio will decrease from 3.5 with decay time to the equilibrium value of 2.2 but cannot be lower. Conversely, if the radionuclides originate from Zircaloy-4, then the ratio would increase with decay time from about 1 to 2.2 but cannot exceed 2.2. Therefore, the observed activity ratios for the Unit 1 (0.1 µm) and Unit 4 (0.45 µm) filters suggest pressure tubes and fuel bundle surfaces, respectively, as the primary source term.
- The Sb-124/Nb-95 ratio for Filters 1 and 2 were 0.022 and 0.092, respectively. These values are consistent with previously reported data for filter particulate [2] and confirm that Sb-124 is generated in particulate form from abrasion between pressure tube and fuel bundle bearing pad surfaces.
- Previous data for a Station-APHT filter, also shown in Table 5, were obtained using a cadmium-zinc-telluride (CZT) detector. Particulars for the Station-A filter were not known. Unlike, the Station-E filters, only the more prominent radionuclides were observed possibly because of the lower efficiency of the CZT detector employed. Comparison of the Station-E and Station-A filter data indicates general consistency, the notable exception being in the case of Co-60. The latter may be partly due to the lower specific activity of Co-60 in Station-A PHT particulate (7E+05 Bq/g [7]) compared to its specific activity in Station-E PHT particulate (3.6E+06 Bq/g [7]).

Table 5: Measured Gamma Activity Results for Station-E Filters

Radionuclide	Station-E Filter 1		Station-E Filter2		Total Station-A Filter Activity (Bq)	Station-A/Station-E Filter Activity Ratio*
	Specific Activity (Bq/cm ³)	Total Activity (Bq)	Specific Activity (Bq/cm ³)	Total Activity (Bq)		
Mn-54	1.2E+05	9.9E+09	1.2E+04	9.3E+08	-	-
Fe-59	1.3E+05	1.0E+10	9.9E+03	8.0E+08	4.0E+08	1/25 (Filter 1) 1/2 (Filter 2)
Co-58	1.4E+04	1.1E+09	1.8E+03	1.5E+08	-	-
Co-60	1.4E+06	1.1E+11	5.6E+05	4.5E+10	2.2E+09	35
Zn-65	4.0E+04	3.2E+09	1.2E+04	9.5E+08	-	-
Zr-95	3.0E+05	2.4E+10	2.4E+05	1.9E+10	8.7E+09	2.5
Nb-94	5.0E+03	4.1E+08	2.2E+03	1.8E+08	-	-
Nb-95	6.9E+05	5.6E+10	4.8E+05	3.8E+10	1.9E+10	2.5
Sn-113	6.9E+03	5.6E+08	4.6E+03	3.7E+08	-	-
Sb-124	1.5E+04	1.2E+09	4.4E+04	3.5E+09	2.1E+09	1.1
Sb-125	3.1E+04	2.5E+09	2.3E+04	1.9E+09	-	-
Cs-134	-	-	8.1E+02	6.5E+07	-	-
Cs-137	6.0E+03	4.9E+08	2.8E+03	2.3E+08	-	-

*Except in the case of Fe-59, the two Station-E filter results were first averaged before calculating the ratio.

6.0 Conclusions

The techniques presented in this paper represent a convenient approach for measuring gamma activity of spent resins and filters. The advantage of gamma scanning spent resins as they are transferred to the storage tank is that the results can be directly correlated to the service conditions of the spent resin column. Sampling resins from the storage tank, while required to develop its full radio-chemical characterization data, has the disadvantage that the identity of the sampled resin cannot be clearly established; because of the associated uncertainty in its transfer date to the storage tank, it is also difficult to adequately decay-correct sample results. In the case of spent filters, gamma scanning at the WWMF, as an alternative approach to the present technique, is logistically complex. In either case, a sample of the filter media (from an ALARA perspective, this is a very challenging undertaking) or PHT system crud is required for complete radionuclide characterization.

The present techniques represent a convenient and relatively inexpensive means for rapidly developing a reliable database for spent resin and filter activities required for the DGR.

7.0 References

- [1]. Y. Verzilov and A. Husain, “Monitoring Of Radionuclide Deposits On CANDU Reactor Components”, Paper presented at 9th CNS International Conference on CANDU Maintenance, Toronto, Ontario, Canada, December 4-6, 2011.
- [2]. A. Husain and Y. Verzilov, “New Insights into Activity Transport Within the Primary Heat Transport Systems of CANDU Reactors”, Paper presented at Nuclear Plant Chemistry Conference, Sapporo, Japan, October 26-31, 2014.
- [3]. A. Husain and D.W. Evans, “Characterization of Ontario Power Generation’s Spent Resins”, Waste Management, Decommissioning and Environmental Restoration for Canada’s Nuclear Activities, Toronto, Ontario, Canada, May 8-11, 2005.
- [4]. A. Husain, “Characterisation of Ontario Power Generation’s Spent Resins”, Paper Presented at Waste Management ’04, Tucson, March 2004.
- [5]. A. Husain, Y. Verzilov and S. Charkov, “In-Situ Gamma Spectrometry of Spent Filter Packages Performed During Unloading Operations into In-Ground Storage Structures at Ontario Power Generation’s Western Waste Management Facility”, Paper presented at EPRI LLW Conference, June 25, 2008.
- [6]. L. Berger, “MERCURAD - Dose Rate Calculation Software”, Canberra Industries, DEX-DT-61316, 2003.
- [7]. A. Husain, V. Chew, R. Taylor, A. Sherlock, “Reference Scaling Factors for Ontario Power Generation’s Low and Intermediate Level Waste: 2005 Update”, OPG Report No 05386-REP-03469.1-00003-R00, December 2005.