Characterization of Sr-90 Drums at the Hanford 618-10 Burial Ground - 16132

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ABSTRACT

The 618-10 Burial Ground on the Hanford Reservation was an early trench waste site used for the disposal of waste from the 300 Area at Hanford. Much of this waste consisted of residues from evaluations of failed fuel as well as various process and separations studies associated with the Department of Energy (DOE) mission which were performed in the 300 Area between March 1954 and September 1963. Historical records indicated that there were also several drums which contained significant quantities of separated Sr-90.

Cleanup of the 618-10 burial ground is covered under the Washington Closure Hanford (WCH) contract. Due to the proximity of the 618-10 site to the Columbia River, waste is being removed from the 618-10 site, stabilized, and disposed of into either the Environmental Restoration Disposal Facility (ERDF) if the containers are determined to be LLW or transferred to the Hanford Central Waste Complex (CWC) if the containers are suspected to be TRU waste. In order to determine the correct disposal path, Non Destructive Assay (NDA) must be performed on all of the containers excavated from the trenches.

The NDA of these containers is performed using germanium gamma spectroscopy with an In Situ Object Counting System (ISOCS) and with a neutron slab counter. Occasional ISOCS spectra exhibited a characteristic Sr-90/Y-90 Bremsstrahlung continuum. In some cases there were measurable gammas present, however in other cases only the Bremsstrahlung continuum was present.

Typically Sr-90 is correlated to Cs-137 as they are normally produced in approximately a 1:1 ratio during the fission process. For some of these drums there was no Cs-137 detectable and therefore no way to quantify the Sr-90. Due to concerns about providing a reasonable and conservative nuclide inventory it was decided to evaluate a technique to utilize data from the Bremsstrahlung continuum to quantify the Sr-90 activity in these drums. The development of the technique combined MCNP modeling and comparison of measured gammas to the continuum for a well-defined waste stream.

The final result showed reasonable correlation to the expected scaling factors for many drums, while also providing a reasonable measured value for drums with significantly enhanced Sr-90 concentrations. Ultimately all drums were demonstrated to be within the ERDF waste acceptance criteria.

INTRODUCTION:

The 618-10 Burial Ground on the Hanford Reservation was an early trench waste site used for the disposal of waste primarily from the 300 Area at Hanford. It accepted waste from March 1954 until September 1963. The 618-10 burial ground contains both trenches, which were used for LLW and some shielded high dose waste drums, and Vertical Pipe Units (VPU) which were used for disposal of non-shielded high dose rate waste. Much of the waste was from evaluations of failed fuel as well as residues from research on various extraction and separations processes. Remediation of the 618-10 burial grounds is a part of the Washington Closure Hanford (WCH) contract. Since the drums cannot be tied to specific historical records, ISOCS measurements are performed to quantify the radioactive content so that the drums can be properly dispositioned. Process knowledge from original records of waste shipments going to 618-10 indicated that there were several drums containing separated Sr-90 or combined Sr-90 and Cs-137. In most waste generated from a fission process Sr-90 is scaled to Cs-137. Normally at 618-10 the scaling factor for the Sr-90 is 1.17 times the Cs-137 activity. However, during routine ISOCS assays it was noted that some drums had a significant Bremsstrahlung spectrum but with little or no measurable Cs-137 present in the spectrum. Sr-90 decays to Y-90, and since both are pure beta emitters, there are no gamma peaks to detect. Therefore Waste Management requested a technique to estimate the Sr-90 content in those drums which obviously did not fit the standard waste profile.

HISTORY:

Historical records indicate that drums containing waste from reactor strontium research had been disposed of in the 618-10 waste site. There are at least 17 drums containing reactor strontium waste with four of those thought to contain purified Sr-90. This waste was disposed in concrete shielded drums, or drums with a combination of lead and concrete shielding.

INITIAL CHARACTERIZATION PROCESS:

Following excavation, the retrieved drum is normally placed into an overpack drum, then sent for characterization. During the initial characterization work a screening was performed using an ORTEC Detective to perform a qualitative identification of the primary nuclides present in the drum. At the same time a qualitative neutron slab measurement using the Hanford Slab Counter (HSC) was performed to check for significant quantities of TRU material. Drums where only U and/or Th was detected did not require further characterization. All other drums were assayed using an ISOCS system for 30 minutes or 2 hours based on whether they were low density debris drums or shielded drums. Of interest relative to the Sr-90 characterization measurement, were shielded drums.

Drums weighing less than 1000 lbs were assumed to be concrete drums and were assayed for 2 hours, to be able to detect transuranic nuclides. Drums which were heavier than 1000 lbs were assumed to be a combination of lead shielding and concrete shielding and were assayed for only 30 minutes since the TRU nuclides would not be detected even in a very long count. The 30 minute count was just to establish the Cs-137 activity. Drums which contained lead shielding had elevated HSC count rates due to spallation of cosmic radiation in the lead shielding creating an elevated neutron flux. The neutron flux was one way of confirming the presence of lead shielding.

In some assays spectra like that shown in Figure 1 were identified.

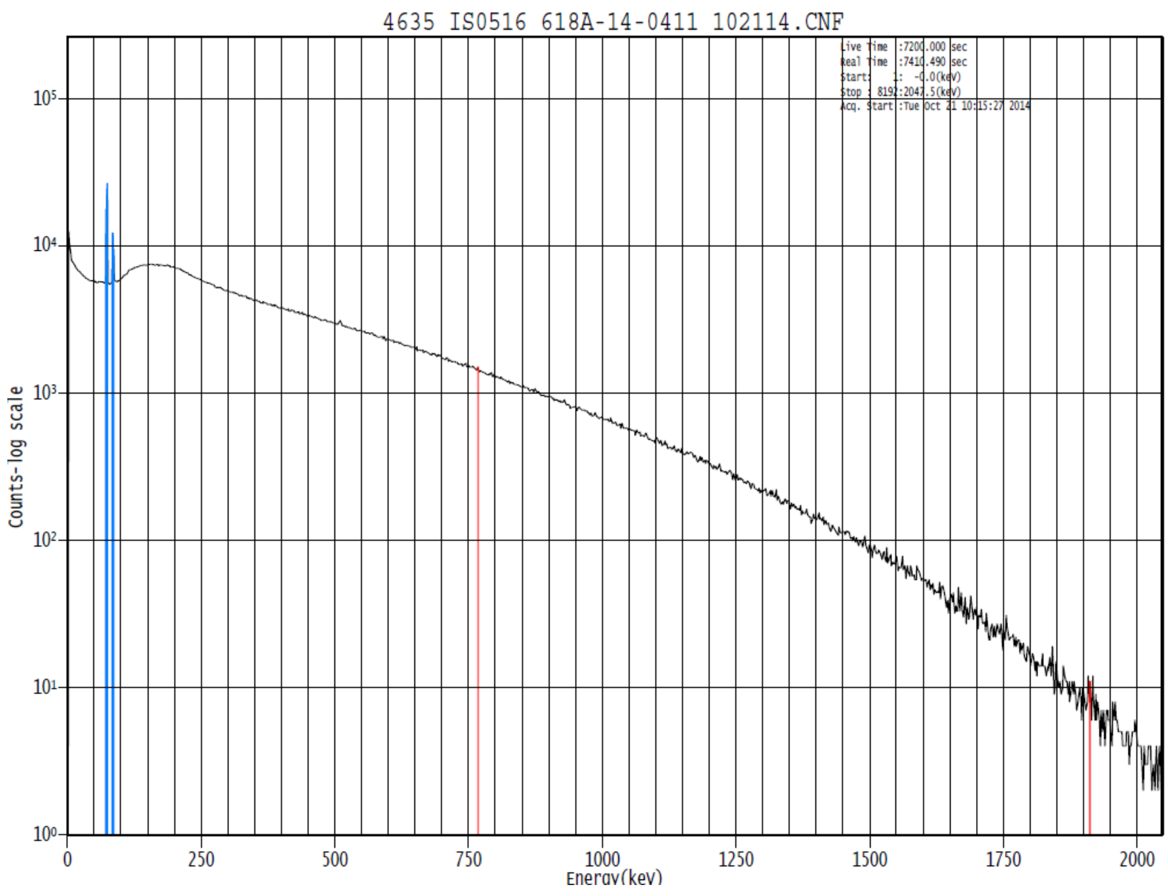

Figure 1: Typical Bremsstrahlung spectrum from Sr-90/Y-90

This spectrum demonstrates a pure Sr-90/Y-90 bremsstrahlung continuum. Since the Cs-137 peak is almost undetectable, traditional gamma spectroscopy techniques cannot be used. Therefore it was decided to study the strength of the bremsstrahlung continuum to attempt to develop scaling factors.

EVALUATION OF PUREX WASTE SPECTRA:

The purest form of fission product waste at the Hanford site was the initial PUREX extraction from the spent fuel. PUREX is the Plutonium URanium EXtraction Process. This separated most of the uranium and plutonium from the fuel leaving the fission products. The scaling factors used for fission product waste at the 618-10 burial ground are based roughly on the PUREX fission product waste stream. Figure 2 is a spectrum of the typical PUREX waste.

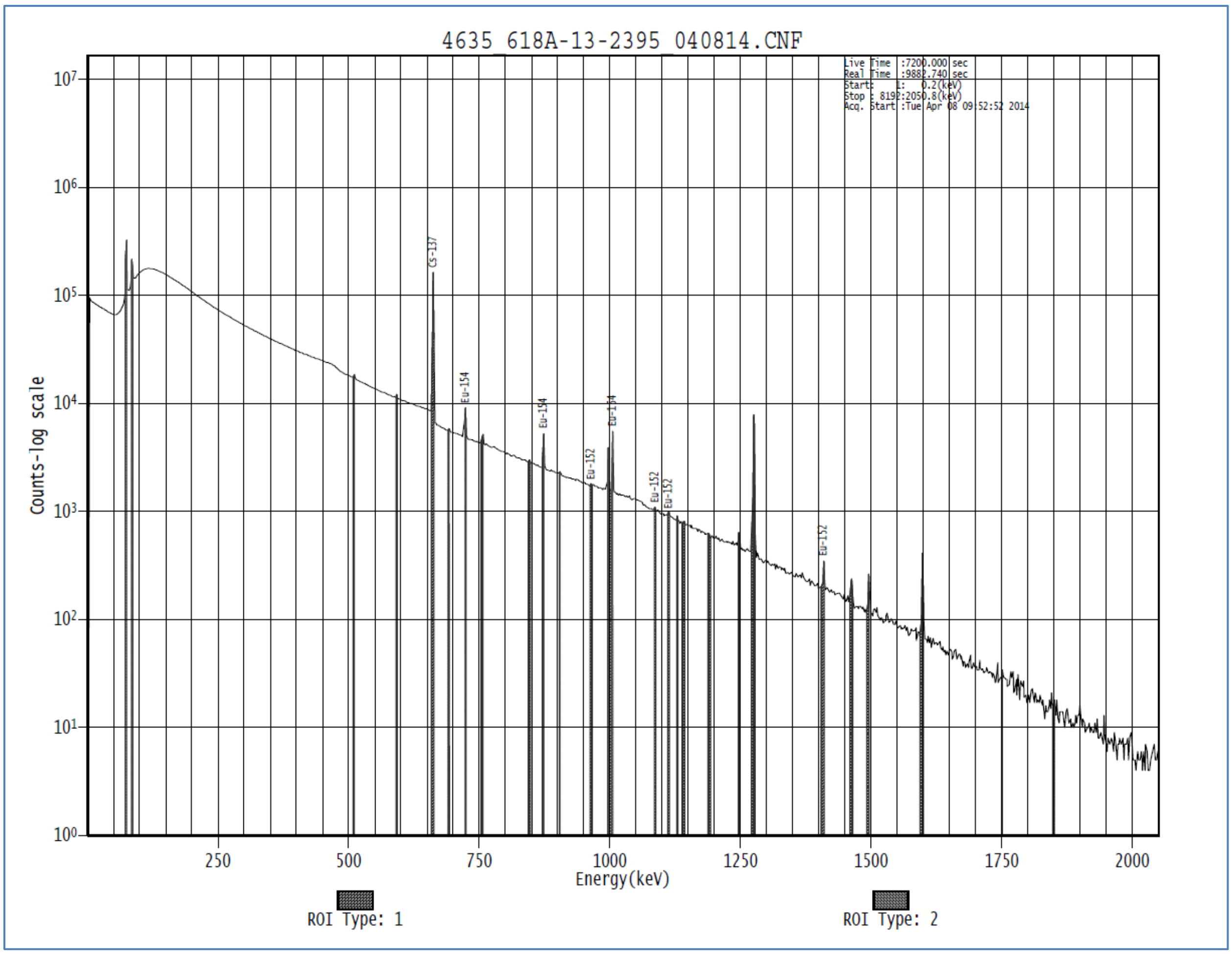

Figure 2: Typical PUREX Fission Product Waste Spectrum in concrete drum

Note that the typical fission products Cs-137, Eu-152, Eu-154 are present, as well as high energy Bremsstrahlung which indicates that Sr-90 is also present. Most other fission products have much shorter half-lives and would be fully decayed. The spectrum in Figure 2 is from a concrete drum assumed to have a 12.5 cm (5”) thick concrete shield. Figure 3 is also a PUREX waste spectrum but in a drum assumed to have a 5cm (2”) lead lining and 15 cm (6”) of concrete shielding.

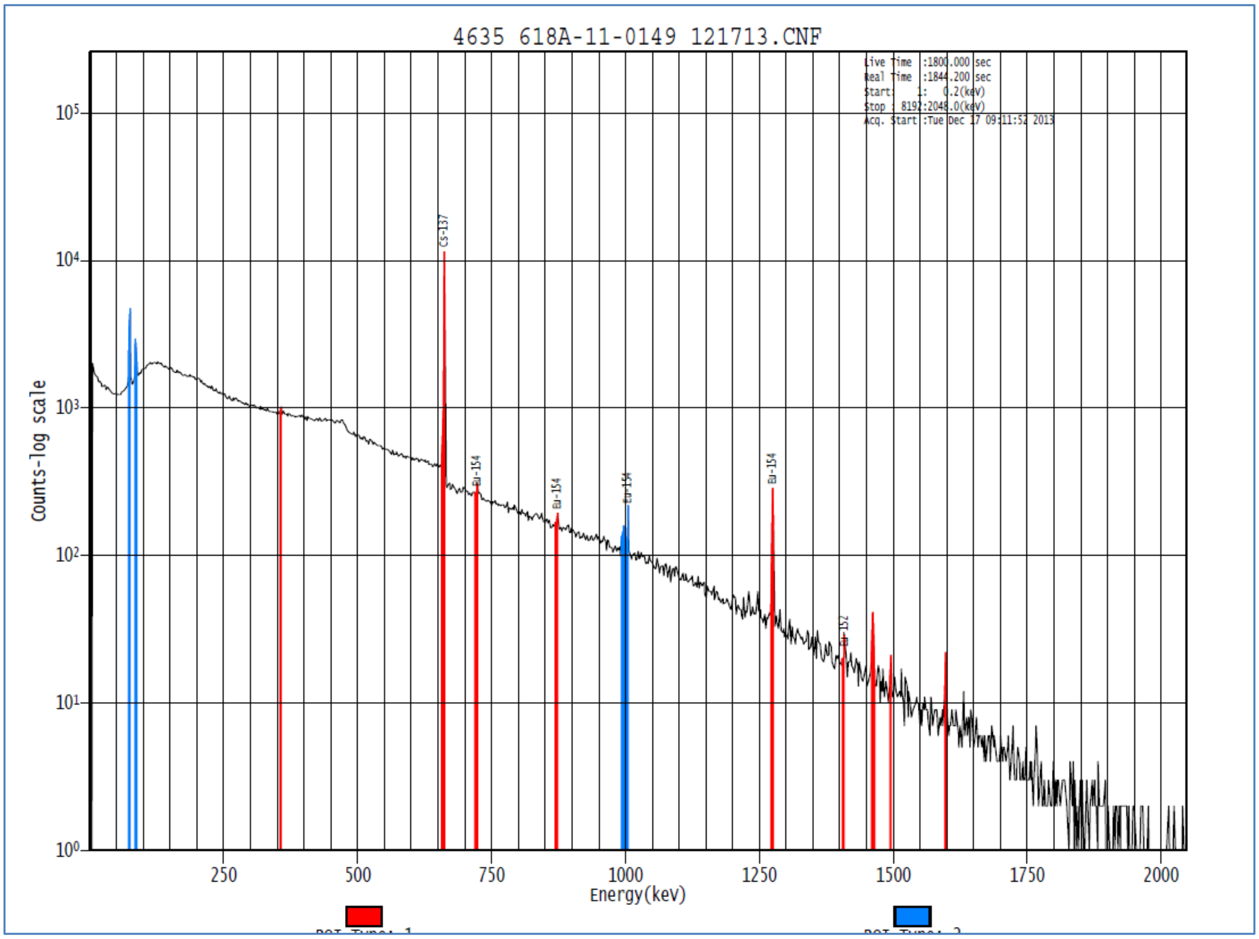

Figure 3: PUREX Fission Product Waste Spectrum in lead lined concrete drum

Since there were a limited number of shielded drums with obvious PUREX waste spectra, four drums were selected. Two of the drums [R-1724, R-1832] had 12.5-15cm (5-6”) thick concrete shields. The other two drums [R-1109, R-1427] had 15cm 6” of concrete shielding and 2.5-5.0cm (1-2”) of lead shielding. The activities for the Cs-137, Eu-152, and Eu-154 were determined from the ISOCS analyses. This was compared to the counts per minute (CPM) from the Bremsstrahlung continuum starting at 680 keV, which is just above the 662 keV peak from the Cs-137. All channels above 680 keV were integrated to generate the CPM value. Occasionally there were other higher energy gamma peaks present, however the contribution from any of these peaks tended to be insignificant in the presence of the Bremsstrahlung continuum and contributed to the conservatism of the calculation. This range was chosen since the Compton scatter from the Cs-137 gamma would significantly affect the number of counts in the continuum below 662 keV, while the peaks and Compton above the 662 keV gamma were typically small and would have a minimal effect on the calculated CPM value. Table 1 shows the results of these measurements.

Table 1: Results for Several PUREX Waste Drums

	Shielding (cm)		Measured Activities (Ci)			CPM	Ratio	Assumed activity (Ci)	CPM/Ci
Drum ID	Conc	Lead	Cs-137	Eu-152	Eu-154	Sr-90	Cs/Sr	Sr-90	Sr-90
R-1109	15	5	7.49E-01	8.43E-05	2.12E-03	1.01E+04	7.44E-05	1.12E+00	1.15E+04
R-1427	16.5	2.5	1.77E+00	MDA	2.45E-04	1.86E+04	9.52E-05	2.07E+00	8.99E+03
R-1724	12.5	0	3.46E-03	1.13E-05	4.05E-04	4.53E+04	7.63E-08	9.56E-03	1.12E+07
R-1832	12.5	0	2.93E-03	MDA	2.79E-05	1.62E+04	1.81E-07	3.42E-03	4.74E+06

Since the calculated CPM per Curie values varied by orders of magnitude between the lead shielded and the concrete drums but only by a factor of 5 for similar shielding configurations it was decided to use one factor for lead+concrete shielded drums and one for concrete shielded drums. The smaller value in each configuration was chosen to provide a more conservative final result for the calculated Sr-90 value. These two factors are shown in Table 2.

Table 2: Sr-90 Scaling factors based on Shielding

Drum shielding (cm)		CPM/Ci
Concrete	**Lead**	**Sr-90**
16.5	2.5	8.99E+03
12.5	0	4.74E+06

RESULTS

Table 3 shows an evaluation of assays which exhibited a Sr-90 Bremsstrahlung continuum. The Sr-90 value calculated based on the Sr-90 continuum and the CPM/Ci factors described in the document is compared to the Sr-90 value developed by scaling to the Cs-137 activity, using the conventional method for this project.

It is interesting to note that most of the values worked out to be within approximately a factor of 5 of each other based on the two techniques. This would tend to indicate that most of the drums are typical fuels waste and therefore fit the standard waste stream profile. The results where the Sr-90 values calculated with this method are significantly higher than scaling to Cs-137 are the drums which contained elevated Sr-90 concentrations. Based on the assumed uncertainties of these calculations, it was assumed that calculated to scaled ratios within a factor of 5 could be assumed to be within agreement. Of the 45 drums in the table, only 17 drums exceeded the cutoff ratio of 5 and a recalculated Sr-90 activity was applied. This is close to the number of enhanced Sr-90 drums which historical records indicated were disposed into the waste site.

Table 3: A comparison of scaled to calculated results for drums of interest

Anomaly #	Drum Wt (kg)	ISOCS Template		Cs-137 Ci	Sr-90 continuum cnts	Measured Sr-90 Activity (Ci)	Sr-90 Concentration (pCi/g)	Sr-90 Based on Scaling Factor	Calc/Scaled
		Conc	Pb						
R-1109	645	15	5	7.49E-01	302200	1.12E+00	1.74E+06	6.21E-01	1.80
R-1427	624	15	2.5	7.65E-02	371591	2.07E+00	3.31E+06	2.07E+00	1.00
R-1724	424	12.5		3.46E-03	5438548	9.56E-03	2.26E+04	4.04E-03	2.37
R-1789	365	10		4.62E-04	397417	6.99E-04	1.91E+03	5.40E-04	1.29
R-1793	395	15		6.22E-04	430224	7.56E-04	1.91E+03	7.27E-04	1.04
R-1799	551	17.5	2.5	9.11E-03	2092003	7.76E+00	1.41E+07	1.06E-02	728.85
R-1800	674	15	5	9.61E-01	502269	1.86E+00	2.76E+06	1.12E+00	1.66
R-1809	442	20		4.96E-03	1383327	2.43E-03	5.50E+03	5.79E-03	0.42
R-1813	462	17.5		3.48E-04	293682	2.07E-03	4.47E+03	4.07E-04	5.08
R-1815	389	0		3.50E-05	280241	2.46E-04	6.33E+02	4.09E-05	6.02
R-1816	583	15		4.20E-03	187331	1.32E-03	2.26E+03	4.91E-03	0.27
R-1818	620	12.5		2.78E-03	159956	1.12E-03	1.81E+03	3.25E-03	0.35
R-1821	497	20		9.25E-06	122627	8.62E-04	1.74E+03	1.08E-05	79.80
R-1822	672	15	2.5	2.51E-03	45160	1.67E-01	2.49E+05	2.93E-03	57.10
R-1824	449	15		3.59E-04	696998	1.23E-03	2.73E+03	4.19E-04	2.92
R-1832	452	12.5		2.93E-03	1945655	3.42E-03	7.57E+03	3.42E-03	1.00
R-1834	491	17.5		2.81E-03	87630	6.16E-04	1.26E+03	3.28E-03	0.19
R-1839	469	17.5		9.31E-04	100282	7.05E-04	1.50E+03	1.09E-03	0.65
R-1841	501	17.5		9.60E-03	201850	1.42E-03	2.83E+03	1.12E-02	0.13
R-1843	619	15	2.5	MDA	6470932	1.20E+01	1.94E+07		
R-1897	421	17.5		8.78E-05	111193	9.77E-05	2.32E+02	1.03E-04	0.95
R-1898	415	20		9.83E-05	137273	1.21E-04	2.91E+02	1.15E-04	1.05
R-1899	404	15		1.03E-05	117028	1.03E-04	2.55E+02	1.20E-05	8.55
R-1906	554	17.5	2.5	MDA	184067	3.41E-01	6.16E+05		
R-1910	405	17.5		3.26E-03	1018070	1.79E-03	4.42E+03	3.81E-03	0.47
R-1911	445	15	5	MDA	1936933	1.80E+00	4.03E+06		
R-1912	398	12.5		1.48E-04	917708	8.07E-04	2.03E+03	1.73E-04	4.67
R-1913	442	15		4.35E-03	3851602	3.57E+00	8.08E+06	5.08E-03	702.56
R-1925	448	5		1.61E-05	139747	1.23E-04	2.74E+02	1.88E-05	6.53
R-1939	462	17.5		5.81E-02	133100	2.47E-01	5.34E+05	6.79E-02	3.64
R-1944	565	12.5		1.83E-02	382630	1.35E-03	2.38E+03	2.14E-02	0.06

R-1946	562	17.5		2.17E-03	194350	1.37E-03	2.43E+03	2.54E-03	0.54
R-1957	443	15		7.81E-05	2287644	4.02E-03	9.08E+03	9.12E-05	44.08
R-1963	612	0		3.63E-04	283007	9.95E-04	1.63E+03	4.24E-04	2.35
R-1972	419	17.5		5.65E-06	66895	1.18E-04	2.81E+02	6.60E-06	17.82
R-1973	409	17.5		4.55E-05	543948	9.56E-04	2.34E+03	5.32E-05	17.99
R-1974	516	12.5		1.19E-04	186348	6.55E-04	1.27E+03	1.39E-04	4.71
R-1985	444	17.5		2.05E-02	197252	9.14E-02	2.06E+05	2.39E-02	3.82
R-1987	384	15		3.82E-05	202035	3.55E-04	9.25E+02	4.46E-05	7.96
R-2263	672	15	2.5	5.65E-02	2808546	5.21E+00	7.75E+06	6.60E-02	78.89
R-2271	629	15	2.5	5.01E-04	140347	2.60e-01	4.14E+05	5.85E-04	444.56
R-2273	503	17.5		8.77E-04	66238	4.66e-04	9.26E+02	1.02E-03	0.45
R-2391	611	15	2.5	MDA	79385	1.47E-01	2.41E+05		
R-2535	407	10		1.40E-06	207100	7.28E-04	1.79E_03	1.64E-06	445.24

CONCLUSIONS:

Although this correction technique was crude it provided a means to provide a reasonable value for Sr-90 in drums which contained enhanced Sr-90 concentrations. This calculated value has proven valuable for the operations group, as the processing of drums containing significant Sr-90 concentrations has had occasional issues with loose contamination. Currently these calculated values are being used to set aside drums from the standard macro-encapsulation process until the contamination issues can be better understood.

A further study of the effects of various drum shields on the Bremsstrahlung continuum was planned but had not been completed in time for the release of this paper. This study is expected to refine the correction factors for the different classes of shielded drums.