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DISCLAIMER

UNDERWATER ANALYSIS OF IRRADIATED REACTOR SLUGS FOR CO-60 AND OTHER RADIONUCLIDES AT THE SAVANNAH RIVER SITE

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ABSTRACT

Co-60 was produced in the Savannah River Site (SRS) reactors in the 1970s, and the irradiated cobalt reactor slugs were stored in the L-Reactor Basin at SRS. Since the activity rates of these slugs were not accurately known, assaying was required. A sodium iodide gamma detector was placed above a specially designed air collimator assembly, so that the slug was eight to nine feet from the detector and was shielded by the basin water. Also, 18 curium sampler slugs, used to produce Cm-244 from Pu-239, were to be disposed of with the cobalt slugs. The curium slugs were also analyzed with a High Purity Germanium (HPGE) detector in an attempt to identify any additional radionuclides produced from the irradiations. Co-60 concentrations were determined for 548 L-Reactor Disassembly Basin cobalt slugs and the 18 curium sampler slugs. The total Co-60 activity of all of the assayed slugs in this work summed to 31,783 curies on 9/15/03. From the Co-60 concentrations of the curium sampler slugs, the irradiation flux was determined for the known irradiation time. The amounts of Pu-238,-239,-240,-241,-242; Am-241,-243; and Cm-242,-244 produced were then obtained based on the original amount of Pu-239 irradiated.

INTRODUCTION

Co-60 was historically produced in the SRS reactors for private vendors and the United States military. Cobalt slugs were irradiated in the early 1970s. Post-production, remaining cobalt slugs (including slab form) were consolidated for storage in L-Basin. There were approximately nine hundred cobalt slugs stored in L-Basin awaiting final disposition. Most of these slugs had historically incomplete documentation for activity rates; therefore, assaying was required in order to determine their activity levels.

Since the gamma dose rate from these slugs is extremely high, the most cost effective way to shield a source of this magnitude from personnel and the radiation detector was to use the basin water in which the slugs were stored as the shield. A sodium iodide gamma detector was placed above a specially designed air collimator assembly, so that the slug was at least eight feet from the detector and was shielded by the basin water.

The assaying took part in two phases. The first phase was in 1999. Assays were taken as slugs were being consolidated from K-Basin to L-Basin and also for a portion of the slugs already stored in L-Basin.[1-3] As these slugs were assayed, they were placed in tag-holders and into labeled

buckets for storage. The second phase of assaying, described in this paper, has been completed for the remainder of slugs that were stored in L-Basin prior to 1999. The slugs that were assayed in the second phase were placed in numbered bins with procedurally specified activity ranges. Only seventy-two slugs had documented activity levels[4] and were not assayed.

All of the slugs previously stored in the basin were disposed of as "low-level waste" at the SRS Solid Waste Facilities. In order to permanently minimize radiation exposure from this intense Co-60 source, a special cask (also slated for waste disposal) was utilized to provide shielding via depleted uranium layers within the cask.

In addition to the cobalt slugs, there were 18 curium sampler slugs[5] (also called Pu monitor pins), used to produce Cm-244 from Pu-239, that were also disposed of in the cask. Very little information existed on the isotopic content of these slugs. Therefore, in addition to the Co-60 assay, the sampler slugs were also analyzed with a High Purity Germanium (HPGE) detector in an attempt to identify any additional radionuclides that remain since the irradiations.

2. EXPERIMENTAL

Instrument Setup for Cobalt Slugs and Slabs

An air filled collimator had been designed to allow a small pencil of gamma radiation from the slug to reach the detector, a 2"x 2" NaI crystal.[2] Nine feet of basin water provided the gamma ray shielding. Figure 1 is a sketch of the collimator used in data acquisition. The collimator consists of a tray to hold the slug that was placed below the lower end of the collimator, a 48"x 3/4" pipe, a 1 1/4" x 48"aperture holder containing five 15/16" x 1/8" aperture pieces and a packet of BBs that dispersed the Co-60 gamma rays. The slug tray was 12" from the pipe cap for the cobalt slug assays, while the aperture pieces and BBs were removed and the tray to pipe cap distance was set at 2 ¾" for the HPGE evaluation of the curium sampler slugs.

Two measurement systems were used for determining the quantity of Co-60 in the cobalt slugs and slabs.[6] For most of the slug and slab measurements, analyses were performed with a portable multichannel analyzer (MCA) EG&G Dart system, containing a laptop computer with GammaVision software. Since the EG&G Dart system became inoperable in the very hot and humid environment, it was replaced with another system that consisted of a portable computer with a Canberra NaI+ card installed and Genie 2000 software. This card converts the PC to a full function MCA and contains the ancillary electronics, high voltage power supply and amplifier, required for data acquisition. A 2" x 2" NaI detector was used and data were stored on floppy disks for subsequent review and analysis and retention. The spectra were acquired in the energy range 0 – 2MeV with an ADC gain of 512 channels and with the detector high voltage set to +800V. Pictures of the NaI detector arrangement and underwater cobalt assay assembly are shown in Figures 2 and 3.

For analysis of the curium sampler slugs, the analysis system consisted of a portable high purity germanium detector, a personal computer with a Canberra Accuspec card and Genie 2000 software, a high voltage power supply, an analog-to-digital converter, and an amplifier.

The gamma ray spectrum from Co-60 consists of two gamma rays, one at 1.17 MeV and the other at 1.33 MeV. A "region of interest" (ROI) was defined around these two gamma rays in the spectrum, and the number of counts in this region summed by the MCA software. This provided the data necessary to perform assays for Co-60.

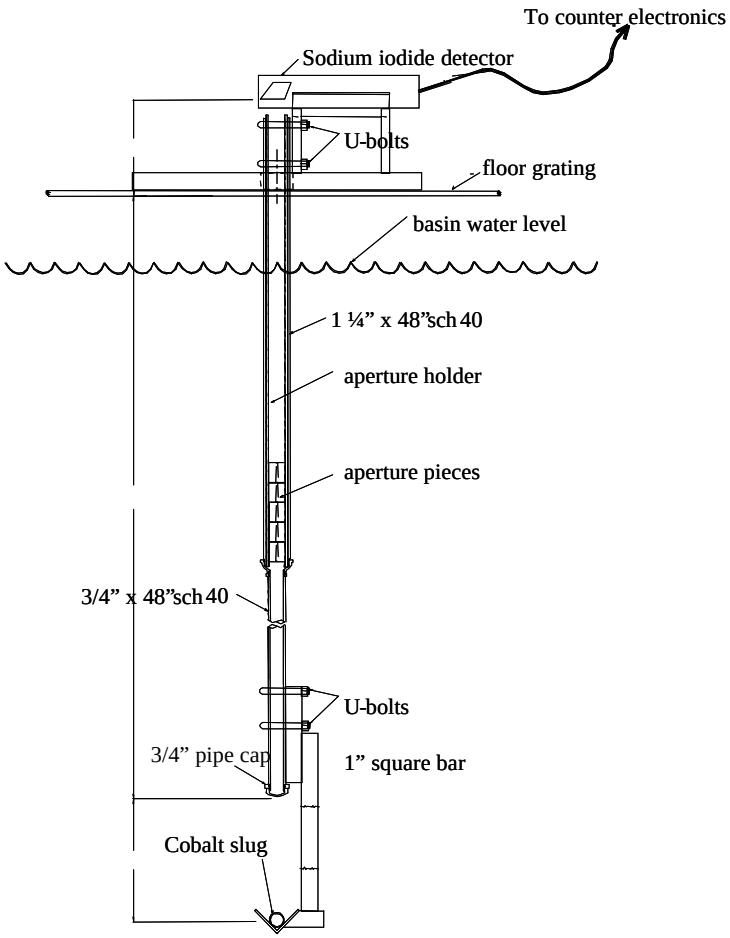

Figure 1. Sketch of the L-Area cobalt assay instrument assembly with NaI detector.

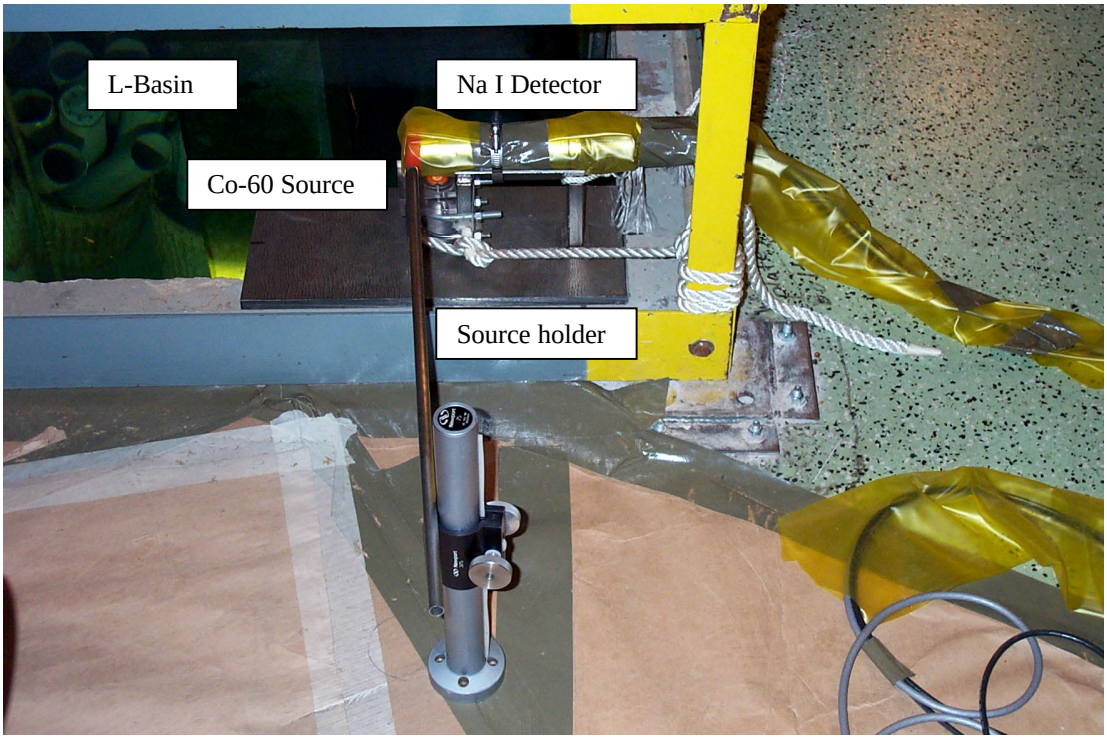

Figure 2. NaI detector arrangement for counting standard Co-60 standard source.

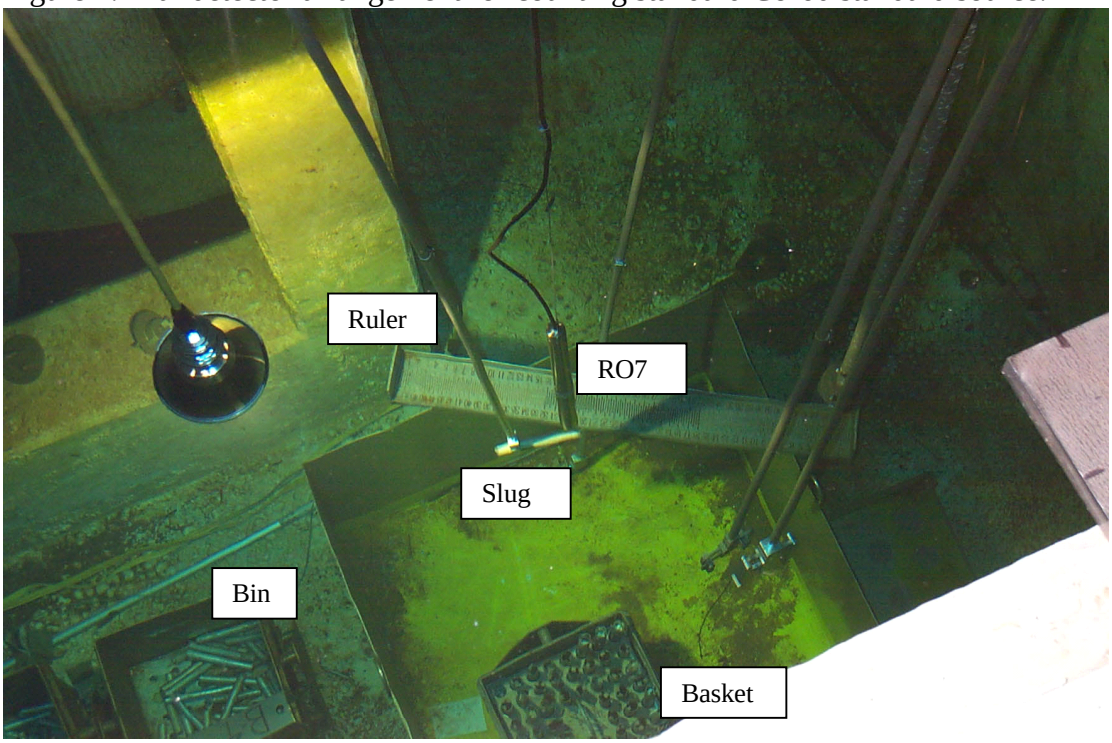

Figure 3. Underwater cobalt assay assembly showing measurement platform, Eberline RO7 monitor and slug bins.

In addition to the cobalt assays, dose measurements were performed, per procedure SOP-DHS-121-L, Assay of Cobalt Slugs,[6] and results were recorded with the assay results. The length of each slug was measured before assays were performed, as shown in Figure 3.

Instrument Setup for Curium Sampler Slugs

In order to increase the sensitivity of the measurement, the sample holder was removed from the water and the slug tray relocated from a distance of 12" to 2 3/4" from the base of the collimator. The small collimators and collimator tube were removed and only the collimator pipe was used.

The 18 curium sampler slugs, used to produce Cm-244 from Pu-239, were removed one at a time from the basket and first assayed with the sodium idodide detector. The Co-60 in these slugs was produced from a 1/16" x 1/16" piece of cobalt wire that was used as a neutron monitor for each irradiation. Since it was impossible to position the slug such that the exact position of the wire was known for this measurement, the Co-60 activity was also obtained from dose readings of the sampler slugs using an Eberline RO7 Middle Range Ion Chamber Survey Meter and by calibrating the RO7 using previously assayed materials.

After the 18 slugs had been assayed with the NaI detector, this detector was replaced with a high-resolution HPGE detector, and the pins were assayed with this detector to determine what isotopes, if any, could be identified in the slugs other than Co-60.

Calibration:

The same calibration was used as in the K Reactor assay described in SRT-ADS-0327, and previous Co-60 slug measurements described in SRT-ADS-99-0391. For this calibration, four slugs were chosen that had different nominal values. The basin background count rate in the ROI was subtracted from the average count and this divided by the average number of curies per slug to obtain the conversion factor of 189 counts/two minutes/Ci for a 2" x 2" sodium iodide detector. The uncertainty in this value was determined to be $\pm$ 25%, and this uncertainty had to be added in quadrature to the other measurement uncertainties, including counting statistics. The slugs in L Reactor Basin are assumed to be essentially identical to those in K Reactor basin.

Both slugs and slabs were analyzed. The slugs have cobalt contained between two D-bar (half-round) pieces of aluminum, while the slabs are pieces (may be rectangular) of cobalt with cladding. Since the gamma rays from Co-60 (1.17 MeV; 1.33 MeV) are very high energy, the difference in sample attenuation for the slugs and slabs is considered well within the overall uncertainties reported. Since the calibration is for a certain length slug (nominally 8") and since gamma rays from only a small portion of the sample are measured, a correction was made for samples of length that differ from the calibration length. The slug tray distance was reduced from 12" to 2 ¾" and the collimation was changed for assays of the curium slugs; therefore, the same slugs were assayed at both configurations to recalibrate the system for the new distance and collimation.

System Quality Assurance/Quality Control:

A background analysis was performed each day prior to sample analysis to show that essentially no Co-60 was present. A Co-60 gamma source was counted in a known geometry, as shown in Figure 2, prior to and after analysis completion to ensure that the energy calibration had not changed significantly and to verify that the counts in a designated peak agree to within control chart limits (3-sigma). All QC checks were within the designated 3-sigma control limits. All of the assay results were independently technically reviewed before the data were released.

3. ANALYSIS AND RESULTS

Cobalt Slug and Slab Analysis and Results

A typical spectrum for the cobalt slugs and slabs is shown in Figure 6. The first region of interest is from Cs-137 in the L-Basin, while the second region of interest is from the two Co-60 gamma rays at 1.17 and 1.33 MeV. The Co-60 activity in this slug was determined to be 72 curies.

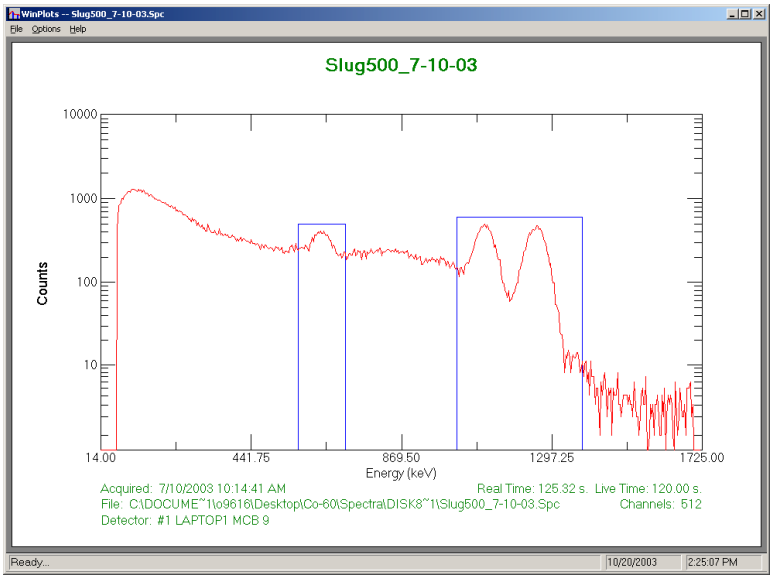

Figure 6. Typical slug spectrum with regions of interest for Cs-137 and Co-60.

The results for this study and previously results are compiled in Table 1. Analysis of 548 slugs. and slabs (items) in this study resulted in 31,783 curies of Co-60 on 9/15/2003. These slugs and slabs were stored in seven bins, and the average item length, dose rate and curies per item are also listed. When previously assayed items were also included for disposal, there were 849 items that contained 126,083 curies of Co-60 on 9/15/2003. These items were placed in a special cask (also slated for waste disposal) utilized to provide shielding via depleted uranium layers within the cask.

Table 1. Co-60 Results for present study and previous work.

Co-60 Results	Bin # (Ci Range)	Items (Slugs+Slabs)	Ave. Length (inches)	Ave. R07 (R/hr @ 24")	Ave. (Ci/Item)	Co-60 (Curies 9/15/03)
Present work	1 (0)	156	7.8	0.3	0	0
Present work	2 (1 - 50)	181	8.5	3.7	22	3957
Present work	3 (50 - 100)	108	9.8	10.6	73	7901
Present work	4 (100 - 150)	32	7.1	13.5	124	3959
Present work	5 (150 - 200)	39	6.9	14.9	177	6885
Present work	6 (200 - 500)	31	7.1	27.2	271	8387
Present work	7 (>500)	1	8.0	48.2	694	694
Total		548				31783
1999		229				61726
Pre-1999		72				32574
All Analyses		849				126083

Curium Sampler Slug Analysis and Results (NaI detector/RO7 Monitor)

The curium sampler slugs had been stored in the basin for over thirty years. They contained a small piece of cobalt wire (1/16" x 1/16") to act as a neutron monitor,[5] and 100 milligrams of Pu-239 to convert to Cm-244. When the slugs were irradiated, the neutron flux converted some of the cobalt into Co-60, which has decayed since the irradiation. By determining the amount of Co-60 produced for a known irradiation time, the neutron flux was determined and the amount of original Pu-239 remaining was determined.[7]

Since the RO7 dose monitor (on contact) measurements were more sensitive for detecting Co-60 than the NaI detector located more than eight feet from the slug, it was calibrated with previously analyzed cobalt slugs and used for the Co-60 determination of the curium slugs.[8]. The curium sampler slugs are composed primarily of aluminum by weight, as compared to the cobalt slugs that are about 48% aluminum and 52% cobalt by weight. Also, the curium sampler slugs are contained in ~1/16" aluminum sleeves, and the cobalt was distributed uniformly throughout the slugs, while the pins only contained a 1/16" x 1/16" wire (neutron monitor). Therefore, dose measurements were adjusted to account for the difference in compositions and cobalt distribution.[9] The measurement uncertainty for the curium sampler slug nuclides contained the slug and RO7 calibrations and the geometry corrections uncertainties and was estimated to be $\pm$35%.

Each of the 18 curium slugs were identical and had essentially identical radiation times. The total radioactivities measured in all of these slugs on 8/4/03 were: Pu-238 (1.1E-04 Ci); Pu-239 (3.0E-02 Ci); Pu-240 (8.9E-02 Ci); Pu-241 (1.7E+00 Ci); Pu-242 (1.4E-04 Ci); Am-241 (2.6 E-03 Ci); Am-243 (4.6E-04); Cm-242 (8.6E-27); and Cm-244 (3.7E-01 Ci).

Curium Sampler Slug Analysis and Results (HPGe detector)

Since a high purity germanium (HPGe) detector has much better resolution than a NaI detector, a 50,000 sec count was done on a sampler slug to search for other gamma-emitting nuclides. In addition to Cs-137 a very small Co-60 peak was detected. No other nuclides were detected below the Cs-137 (661.65 keV) peak because of the high Compton background from Cs-137 in the basin. In addition to Cs-137, the only other discernable peaks detected were for the nuclides Eu-154 (723.4; 873.5; 1004.9;1274.3 keV), Co-60 (1173.2; 1332.5 keV) and K-40 (1460.6 keV).

CONCLUSIONS

Using a sodium iodide detector and multichannel analyzer system and a specially designed underwater collimator assembly, the Co-60 concentrations were determined for 548 L-Reactor Disassembly Basin cobalt slugs and slabs (items) and 18 curium sampler slugs. The total activity of the 548 slugs and slabs assayed in this work summed to 31,783 curies (9/15/03). No Co-60 was detected for 156 items, while 181 averaged 22 curies; 108 averaged 73 curies; 32 averaged 124 curies; 39 averaged 177curies; 31 averaged 271 curies, and 1 had 694 curies.

Since the amount of Co-60 in the curium sampler slugs was not detectable with the sodium iodide detector, it was determined by calibrating an RO7 Ion Chamber Survey Meter with a slug of known activity. A geometry correction was made for the curium sampler slug cobalt distribution (1/16" x 1/16' wire) compared to the cobalt slug geometry (8" rod). From the Co-60 of the curium sampler slugs, the irradiation flux was determined for the known irradiation time. The amounts of Pu-238,239,240,241,242; Am-241,243; and Cm-242,244 produced were then obtained based on the original amount of Pu-239 irradiated. The curium sampler slugs were analyzed with a high purity germanium detector (50,000 sec). In addition to Cs-137 from the basin water, the only other discernable peaks were from Co-60, Eu-154 and K-40.

6. REFERENCES

1. F. S. Moore and Saleem Salaymeh, WSRC Memorandum SRT-ADS-99-0327, Co-60 *Assay Values of Co-60 Slugs in K Reactor Disassembly Basin*, 8/10/99.
2. Frank Moore and Saleem Salaymeh, WSRC Memorandum SRT-ADS-99-0391, *Co-60 Assay Values of Co-60 Slugs in L Reactor Disassembly Area*, 9/13/99.
3. Frank Moore and Saleem Salaymeh, WSRC Memorandum SRT-ADS-99-0453, *Assay of Miscellaneous Slugs in L Reactor Storage Basin*, 11/3/99.
4. WSRC Procedure SOP-DHS-117-L, Rev. 0, *Receipt of Cobalt in 8-Ton SRL Cask-Disassembly*, 6/03.
5. Hilley, J.R., Smith, J.A., Nelson, E.C., *Curium-244 Production Program*, WSRC Technical Report TA 1-1361, 4/21/64.
6. WSRC Operating Procedure SOP-DHS-121-L, Rev. 0, *Assay of Cobalt Slugs*, 6/03.
7. MacLeod, S.M., *Curium-244 Production Program*, WSRC Technical Report TC 1-1361, 10/65.
8. Casella, V.R., *Determinations of Radionuclides in L-Area Curium Sampler Slugs (Pins)*, WSRC Engineering Calculation G-GLC-A-00116, 9/16/03.
9. Microshield v5.03 (5.03-00075), Grove Engineering, Rockville, Maryland, 10/96.