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TUNGSTEN SHIELDS FOR CS-137 INLINE MONITORS IN THE SRS CAUSTIC SIDE SOLVENT EXTRACTION PROCESS

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The Department of Energy (DOE) selected Caustic-Side Solvent Extraction (CSSX) as the preferred technology for the removal of radioactive cesium from High-Level Waste (HLW) at the Savannah River Site (SRS). The CSSX process is a continuous process that uses a novel solvent to extract cesium from highly radioactive waste and concentrate it in dilute nitric acid. Inline analyses are performed with gamma-ray monitors to measure the C-137 concentration in the decontaminated salt solution (DSS) and in the strip effluent (SE). Sodium iodide (NaI) monitors are used to measure the Cs-137 concentration before the DSS Hold Tank, while Geiger-Mueller (GM) monitors are used for Cs-137 measurements before the SE hold tank. Tungsten shields were designed using Monte Carlo calculations and fabricated to provide the needed reduction of the process background radiation at the detector positions. A one-inch tungsten cylindrical shield reduced the background radiation by a factor of fifty that was adequate for the GM detectors, while a three-and-one-half-inch tungsten cylindrical shield was required for the NaI detectors. Testing of the NaI shield was performed at the SRS Instrument Calibration Facility. Based on this testing, the as-built shield is predicted to be able to detect the MCU DSS stream at concentrations above 0.003 Ci/gal under the "worst case" field conditions with a MCU feed solution of 1.1 Ci/gal and all of the process tanks completely full. This paper discusses the design, fabrication, testing and implementation of the tungsten shields in the MCU facility.

INTRODUCION

The Department of Energy (DOE) selected Caustic-Side Solvent Extraction (CSSX) as the preferred technology for the removal of radioactive cesium from High-Level Waste (HLW) at the Savannah River Site (SRS). Before the full-scale Salt Waste Processing Facility becomes operational, a portion of dissolved saltcake waste will be processed through a modular CSSX unit (MCU). The MCU employs the CSSX process, a continuous process that uses a novel solvent to extract cesium from waste and concentrate it in dilute nitric acid. Of primary concern is Cs-137 which makes the solution highly radioactive. Since the MCU does not have the capacity to wait for sample results while continuing to operate, the Waste Acceptance Strategy is to demonstrate compliance to downstream facility Waste Acceptance Criteria by performing inline analyses. Gamma-ray monitors are used to measure the C-137 concentration in the decontaminated salt solution (DSS) and in the strip effluent (SE). Sodium iodide (NaI) monitors are used to measure the Cs-137 concentration before the DSS Hold Tank, while Geiger-Mueller (GM) monitors are used for Cs-137 measurements before the SE Hold Tank. The Cs-137 concentration in the DSS Hold Tank will be very low (<0.1 Ci/gal) and the monitor will be in a very high radiation area (up to 4 rem/hr). Therefore, the NaI detector shield design is a critical component in ensuring that the facility background is reduced sufficiently so that an accurate measurement can be performed. The Cs-137 concentration in the SE Hold Tank is about fifteen times higher than the Cs-137 concentration in the Feed Tank (<16.5 Ci/gal), and the GM detector tungsten shield must reduce the facility background radiation by about a factor of fifty. Tungsten shields were designed using Monte Carlo calculations and fabricated to provide the needed reduction of the process background radiation at the detector positions.

MEASUREMENT PROCESS

NaI Detectors

Irradiations were done at the Savannah River Site Calibration Facility using a high-activity Cs-137 source that provided beam strengths of 3.27 to 3.48 rem per hour. A thallium-doped sodium iodide, NaI (Tl), gamma-ray detector with personal-computer-based data-acquisition electronics and spectral stripping software provided a measured count rate in the region-of-interest.

The tungsten shield was tested at the calibration facility and the results were converted to those that would be expected at the MCU facility. At the MCU facility, repetitive 30-minute count times are presently planned and 30-minute counts times were also used at the test facility. The detector-shield configuration was calibrated with specially ordered NIST-traceable pipe standards that consist of MCU piping and Cs-137 contained in an epoxy material with a density closely matched to the process solution. The dose rates used at the calibration facility are comparable to the MCU dose rates (3.66 to 3.99 rem per hour) for the bottom, South and West shield directions. The MCU dose rate for the top of the shield is lower (1.35 rem per hour) and for the East and North shield directions the MCU dose rates are negligible. The testing takes into account these dose rate differences.

The 662-keV beam flux at the test facility is much more penetrating than the MCU beam flux that has a range of gamma energies (0 to 662 keV) due to Compton Scattering. A correction of the data for this difference in flux energies is also made based on MCNP calculations. Figure 1 shows the preparation of the shield for irradiations and Figure 2 shows an example of the NaI spectrum (not from the test facility) while Figures 3 and 4 show the vertical and horizontal irradiation positions.

The test configurations are given in Procedure SP-17-010 Rev.1, “MCU Tungsten Gamma Shield Performance Test.” The irradiation positions of the shield were:

- Shield vertical (rotate every 30 degrees for 12 irradiations)
- Shield top and bottom (irradiate top and bottom and rotate 30 degrees in each

direction for 6 irradiations)

Figure 1. Shield preparation for irradiations.

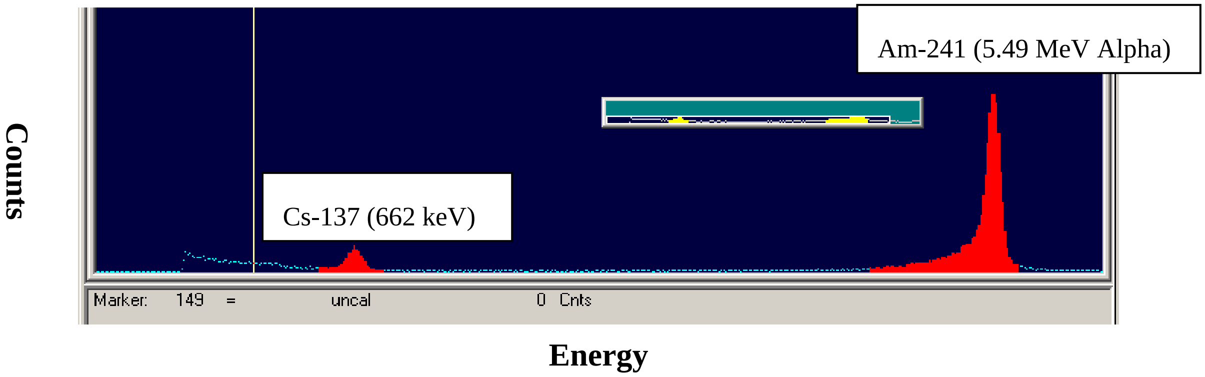

Figure 2. Spectrum from Na(Tl) detector with Am-241 Pulser

Figure 3. Tungsten shield in vertical position.

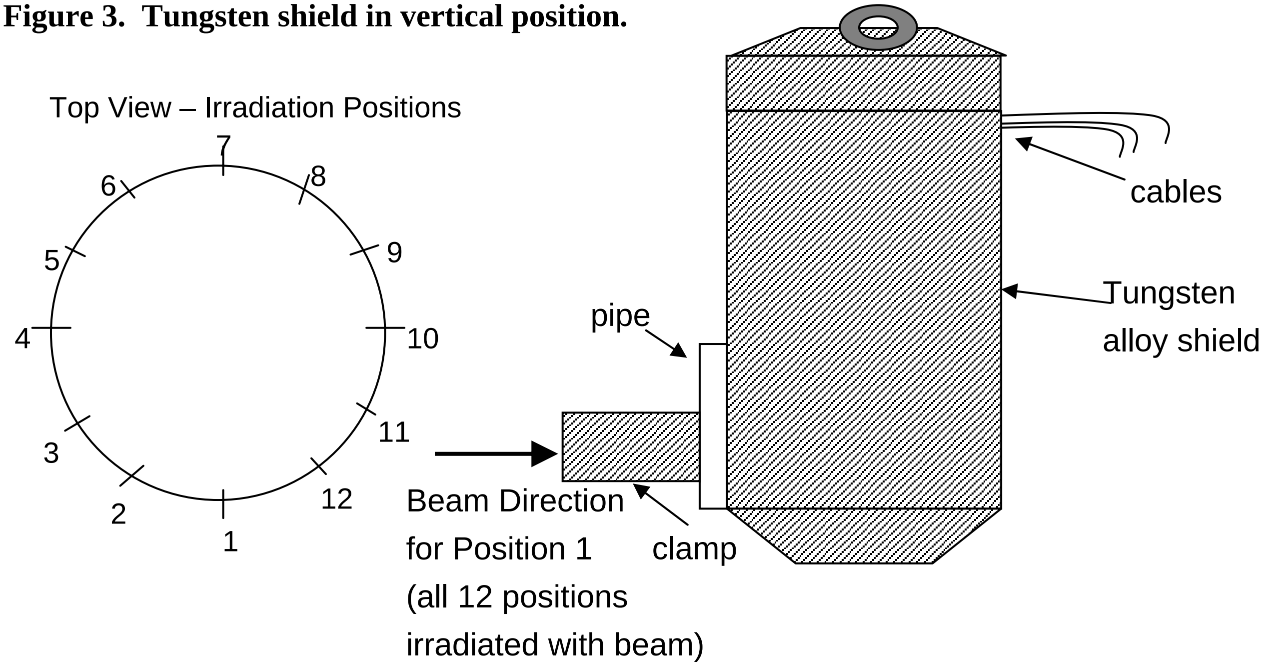

Figure 4. Tungsten shield in horizontal position.

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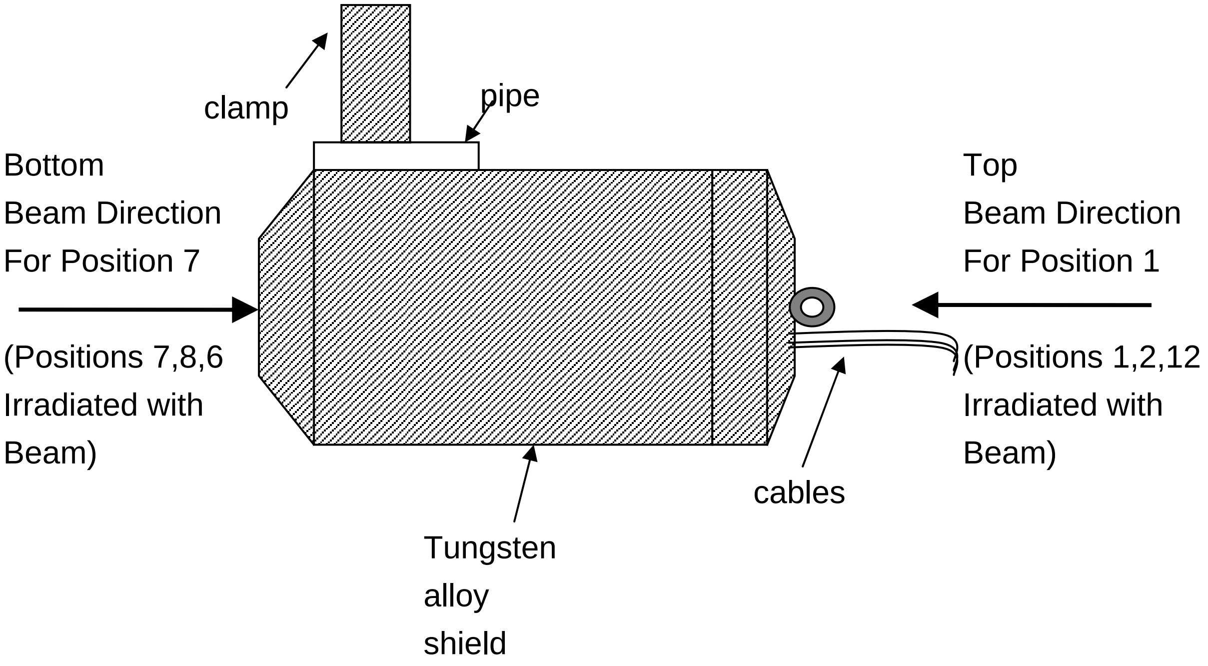

The results for the test irradiations are presented in Table 1.

Table 1. Results for 30-minute Irradiations of West Detector.

Position	Beam Rem/Hr	Cs-137 Peak (c)	Cs-137 Uncert. (c)	Cs-137 Integral (c)	Cs-137 MDA (c)	Cs-137 MDA or Peak (cps)	Am-241 Integral (c)
Vertical							
1 (West)	3.40	280	107	4828	323	0.179	840742
2	3.40	0	0	4641	317	0.176	839706
3	3.40	0	0	4947	327	0.182	839755
4 (North)	3.40	115	109	4974	328	0.182	841185
5	3.40	197	110	5027	330	0.183	843089
6	3.40	229	110	5076	331	0.184	843337
7 (East)	3.40	0	0	4846	324	0.180	844252
8	3.40	32	110	4948	327	0.182	846138
9	3.40	0	0	5066	331	0.184	845704
10 (South)	3.40	349	109	5070	331	0.194	845652
11	3.40	121	110	5049	330	0.184	842101
12	3.40	277	102	4457	310	0.172	842404
Horizontal							
7 (Bottom)	3.27	2100	133	8477	428	1.167	843955
8	3.31	0	0	4925	326	0.181	841554
6	3.31	0	0	4637	317	0.176	844021
1 (Top)	3.48	693	109	5276	338	0.385	846963
2	3.44	0	0	4318	306	0.170	846390
12	3.44	0	0	4410	309	0.172	844581

The following conclusions can be derived from the data given in Table 1. The last column (Am-241, counts) shows extremely consistent results for the Am-241 peak for each measurement. This peak is due to the Am-241 “seed” on the detector itself and will give essentially the same results if the detector is working properly. These results validate that the detectors and electronics were operating properly throughout the test.

The first column gives the irradiation positions previously discussed; the second column gives test beam intensity in rem per hour; the third and fourth columns give the measured Cs-137 peak and uncertainty counts; the fifth column is the integral counts in the Cs-137 spectral region-of-interest; the sixth column is the minimum detectable activity in counts calculated from the integral counts; the seventh column is the larger of the measured peak area or MDA for each position divided by the count time of 1800 seconds. The Cs-137 peak refers to the stripped result (background under peak subtracted), while the Cs-137 integral contains all counts in the region-of-interest.

In the listed positions, except positions 10 (South), 7 (Bottom) and 1 (Top), the measured peak area is less than the minimum detectable activity (MDA). Therefore, there are no discernable Cs-137 gamma rays that penetrate the shield for these positions. Position 10 (South) is barely above the MDA, while the only statistically significant Cs-137 peaks observed were for positions 7 (Bottom) and 1 (Top). This might be expected for the top because the eye hook removes some of the tungsten shielding directly above the detector and replaces it with steel. The top chamfer and cable groove would not provide a reduction in tungsten thickness for the top irradiation because they are not in line with the direction of the test beam. The irradiations at ± 30^0 from the top (positions 2 and 12) were also not affected by this chamfer and groove as shown by the experimental results.

However, it is not apparent why there is a small amount of Cs-137 gamma rays detected when the beam irradiation is directly on the bottom of the shield. It would appear that there is slightly less shielding on the bottom of the shield. Ultrasonic testing did not detect a void in the tungsten at the bottom of the shield. In the MCU configuration, there is also an I-beam beneath the shield that provides approximately one inch of steel as additional shielding and would reduce this contribution by at least a factor of three. As shown by subsequent discussion, these small observed Cs-137 peaks from the test beam have only a very small contribution to the shield MDA.

GM Detectors

MCNP calculations showed that a one-inch diameter tungsten alloy shield was adequate for the GM detectors and the design is given in Figure 5. The tungsten clamp also provided a means to insert a detector calibration source. This shield reduced the background radiation by a factor of 48; therefore, only about 98 percent of the background radiation was removed. For the detectors used in the MCU process, this will represent less than a 5 percent increase in background radiation. A correction can be made for the increase in background, and the overall uncertainty will be well within the process requirement of ± 20%.

Figure 5. GM tungsten shield and calibration source holder.

DATA EVALUATION

Several experimental parameters must be considered in order to calculate the shield detection limit. Some of these parameters are given in Table 2 for the four shield positions that contribute to the detection limit. In order to equate the test beam to the MCU radiation field, the beam dose rate (Rem/hr) must be known for each facility, and a factor must be used to account for the difference in gamma ray energies for the two beams. These parameters are given in Table 2. The beam energy factor of 3.09, obtained from MCNP calculations, means that the test facility 662 keV beam is 3.09 times more likely to penetrate the shield than the MCU radiation field in the detected range of 580 to 700 keV. (Report N-CLC-H-00672 gives a factor of 3.64 for gamma rays detected for a 4 Rem/hr test beam and the MCU average dose rate of approximately 3.4 Rem/hr. It is reduced to 3.09 to equate these dose rates.)

The Cs-137 Integral (21.6 ±0.2 cps) is the counts per second for a measured 0.01 Ci/gal solution as determined by a pipe standard measurement in the exact MCU configuration. This agrees to within the counting uncertainty of the value of 21.8 cps predicted by the MCNP calculations (N-CLC-H-00672) based on previous standard measurements. This column is particularly noteworthy because it gives the count rate for gamma rays in the Cs-137 region-of-interest and tailing from the Am-241 peak. Therefore, it is used for calculation of the detection limit. As stated, the Cs-137 integral count rate for a 0.01 Ci/gal solution in the process piping is 21.6 cps; however, the background requirement is 10% of this value. Since the MCU background radiation changes as the process changes (feed solution concentration, amount of solutions in the tanks, etc.), this 10% requirement allows adjustment for any background radiation that penetrates the shield. This is a small correction that would still be within the overall measurement uncertainty of 10%.

Table 2. Data for Detection Limit Calculation.

Position	Test Beam Rem/hr	MCU Beam[1] Rem/hr	Beam Energy Factor (Test/MCU)	Cs-137 Integral cps/0.01 Ci/gal	DL Required = 0.1 x 21.6 cps/0.01 Ci/gal
Top	3.27	1.35	3.09	21.6	2.16
Bottom	3.48	3.66	3.09	21.6	2.16
South	3.40	3.80	3.09	21.6	2.16
West	3.40	3.90	3.09	21.6	2.16

[1] The dose rates (Rem/hr) of the East and North sides of the tungsten shield are negligible.

Table 3 lists the beam corrected count rates and detection limits (Ci/gal) calculated for the top, bottom, South and West directions of the tungsten shield configuration in the

MCU. The calculation formulae used for this calculation are given in the table footnote.

Table 3. West Detector Detection Limit for a 30-min Count for 1.1 Ci/Gal Feed Solution and Process Tanks Full of Solution.

	Cs-137 MDA or Peak cps	MCU Shield cps	Detection[1] Limit (Ci/gal)
Top	0.385	0.051	0.00024
Bottom	1.167	0.397	0.00184
South	0.194	0.070	0.00032
West	0.179	0.066	0.00031
	Total =	0.59	0.0027

[1] MCU Shield (cps) = (cps Cs-137)(MCU Beam (Rem/hr) / Test Beam (Rem/hr))(1/3.09)
[2] DL (Ci/gal) = MCU (cps) x (0.01 Ci/gal / 2.16 cps)

As shown in Table 3, the tungsten shield MDA for a 30-min count time and the "worst case" facility conditions of having 1.1 Ci/gal feed solution and having all of the process tanks full is determined to be ≤0.003 Ci/gal. The primary factors contributing to the detection limit are a very small amount of Cs-137 activity detected through the shield bottom and the Am-241 background tailing observed in the Cs-137 region-of-interest.

Since the steel I-beam below the shield will provide attenuation of at least a factor of three, the "worst case" shield MDA will actually be <0.002 Ci/gal because the MDA of the bottom would be reduced by 0.0012 Ci/gal. If only the tailing from the Am-241 peak is considered for the present NaI detectors that contain a 27 nCi of Am-241, then the lowest MDA that can be achieved, based on the sum of the lowest possible MDAs (0.00011; 0.00028; 0.00032, and 0.00031 Ci/gal for the Top, Bottom, South and West , respectively), is 0.001 Ci/gal for a 30-min count time. About 27 nCi of Am-241 was used for the detector pulser, while 10% of this amount is adequate to obtain excellent counting statistics for the pulser peak. When additional detectors are used, the Am-241 seed should be in the range of 2 to 5 nCi to lower the shield detection limit even more.

Based on this experiment, the as-built shield is predicted to be able to detect the MCU DSS stream at concentrations above 0.003 Ci/gal under the "worst case" field conditions. If the feed solution radioactivity concentration decreases, a corresponding decrease will be observed for this detection limit (to a lower limit of 0.001 Ci/gal for the 27 nCi Am-241 seeded detectors presently used.). This experiment showed that the facility radiation not attenuated by the tungsten shield would represent less than a 3% increase of the measured background radiation. Since this would result in a very slight conservative bias in the measurement and since the required measurement accuracy is ±10%, the slight increase in background can be ignored or a very small correction can be used.

CONCLUSIONS

- The as-built tungsten shield will be able to detect the MCU DSS stream at concentrations above 0.003 Ci/gal under the "worst case" field conditions.

- The tungsten shield was irradiated in 12 vertical (every 30 degrees) and 6 horizontal positions (top and bottom) and there was no Cs-137 peak observed above the MDA for 16 of these positions. Very small Cs-137 peaks were observed when the beam was directly on the top or bottom of the shield, but no peaks were observed at angles of ±30 degrees of the top and bottom. The top beam penetration may be because of the removal of tungsten shielding for the eye hook. It is unclear why there is beam penetration on the shield bottom but this will be mitigated at least a factor of three by the steel I-beam used for support.

- This experiment showed that there is excellent agreement between the experimental results and the MCNP calculations based on previous standard measurements. The MCNP calculations predicted that, based on the collimator diameter and length, the Cs-137 integral counts would be 21.8 cps and the measured value was 21.6 ± 0.2 cps.

- The slight increase in background due to the radiation penetrating the shield can be ignored or a very small correction can be used. Since it is unlikely that the actual "worse case" conditions will be approached, it is not likely that a background correction will be necessary. If the "worst case" conditions were approached, a prudent background correction between 1 to 2% of the MDA (21.6 cps for the present detectors) can be made based on the present work.

- When additional detectors are used, the Am-241 seed should be in the range of 2 to 5 nCi to lower the Am-241 tailing and detection limit in the Cs-137 region-of-interest .

- If only the tailing from the Am-241 peak is considered for the present NaI detectors that contain a 27 nCi of Am-241, then the lowest MDA that can be achieved is 0.001 Ci/gal for a 30-min count time. A lower detection limit can be achieved by increasing the count time or using detectors with less Am-241 seed. A two-hour count time will reduce the detection limit by a factor of two.

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