Characterization of Filters Loaded With Reactor Strontium Carbonate - 13203

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ABSTRACT

A collection of three highly radioactive filters containing reactor strontium carbonate were being prepared for disposal. All three filters were approximately characterized at the time of manufacture by gravimetric methods. The first filter had been partially emptied, and the quantity of residual activity was uncertain. Dose rate to activity modeling using the Monte-Carlo N Particle (MCNP) code was selected to confirm the gravimetric characterization of the full filters, and to fully characterize the partially emptied filter.

Although dose rate to activity modeling using MCNP is a common technique, it is not often used for bremsstrahlung-dominant materials such as reactor strontium. As a result, different MCNP modeling options were compared to determine the optimum approach. This comparison indicated that the accuracy of the results were heavily dependent on the MCNP modeling details and the location of the dose rate measurement point. The optimum model utilized a photon spectrum generated by the Oak Ridge Isotope Generation and Depletion (ORIGEN) code and dose rates measured at 30 cm. Results from the optimum model agreed with the gravimetric estimates within 15%.

It was demonstrated that dose rate to activity modeling can be sucessful for bremmstrahlung-dominant radioactive materials. However, the degree of success is heavily dependent on the choice of modeling techniques.

INTRODUCTION

Hot cell cleanout activities at the High-Level Radiochemistry Facility (HLRF) necessitated the expedited disposal of three highly radioactive filters containing reactor strontium carbonate left over from a legacy medical isotope campaign. To facilitate the disposal effort, the quantity of Sr-90 in the filters had to be definitively established. Dose rate measurements and MCNP modeling were used to confirm earlier gravimetric estimates for all three filters.

HIGH LEVEL VAULT CLOSURE PROJECT

The High Level Vault (HLV) is an underground shielded strucutre within the boundary of the Hanford 324 Building that houses four stainless steel tanks. The tanks in the HLV were used to store liquids associated with research in the 324 Building hot cells, including spent reactor fuel reprocessing and high-level waste vitrification efforts. Upon completion of the research missions, these tanks together contained approximately 480 TBq (13,000 Ci) of Sr-90 and 740 TBq (20,000 Ci) of Cs-137, with varying levels of chemical and radioactive impurities.

As part of the effort to close the HLV, the decision was reached to recover Sr-90 and Cs-137 from the HLV tanks [1]. Recovery of the Sr-90 and Cs-137 was intended to simplify the management of the remaining liquid waste, with an added benefit of producing a store of purified Sr-90 to supplement existing medical isotope stocks.

The liquid waste in the HLV tanks was first neutralized, and the resulting precipitates were removed by filtration. The Sr-90 was removed by carbonate precipitation/filtration and the Cs-137 was removed by

ion exchange. The remaining liquid was then evaporated, and the solid wastes were disposed through existing pathways.

A total of five filters loaded with Sr-90 carbonate were produced during processing of the HLV tank liquids. However, only the first three filters were saved for future use due to the presence of significant levels of alpha producing transuranics in the feed for the last two filters.

The initial and final weight of each filter was used to determine the mass of carbonate. The chemical composition of the feed, estimated processing efficiencies and an assumed composition of reactor strontium were then used to convert the mass of carbonate into the mass of Sr-90. The first filter was estimated to contain 67 TBq (1800 Ci) of Sr-90. The second and third filters were estimated to contain 140 TBq (3800 Ci) of Sr-90 each [2].

MEDICAL ISOTOPE PROGRAM

Monoclonal antibodies directed against tumor-specific antigens, when labeled with beta-emitting radioisotopes such as Y-90, have proven to be effective for the treatment of several classes of human tumors that are relatively unresponsive to conventional chemotherapy or external beam radiotherapy [2]. Such antibodies represent the primary use of medical grade Y-90.

The Pacific Northwest National Laboratory (PNNL) has long been a major source of medical-grade Y-90 for the United States and Europe. Y-90 is generated by radioactive decay of Sr-90, and when a sufficient amount has collected it may be separated from the parent by solvent extraction. Since the separated Y-90 has a half-life of 64.1 days it must be used quickly and replenished regularly [3].

To meet continued growth in the demand for Y-90, PNNL undertook a campaign in 2000 - 2003 to purify an additional 55.5 TBq (1500 Ci) of Sr-90 to serve as a Y-90 generator. The filters from the HLV closure project were relocated to the HLRF to support Sr-90 recovery and additional purification. The lowest activity filter of the three was used as a feedstock for this process [2]. The other two filters were retained for future use.

The strontium carbonate was dissolved using 2M nitric acid and the solution was removed from the HLV filter casings. Metal oxides were again precipitated and removed, and the Sr-90 was re-precipitated as a carbonate and collected in filters of a new design. Cs-137 impurities were removed by repeated washing. Chemical analysis indicated that the final product was acceptable for the medical isotope program. The chemical analysis also indicated that the ratio between calcium and strontium used in the earlier gravimetric estimates of the HLV filters yielded results that were approximately 20% too high [2].

The estimated Sr-90 content in the filters is shown in Table I.

Table I. Sr-90 Filter Activity Estimates

Filter	Initial Estimate	After Ca/Sr Ratio Adjustment	After Sr-90 Recovery
Sr-01	1800 Ci	1440 Ci	?
Sr-02	3800 Ci	3040 Ci	3040 Ci
Sr-03	3800 Ci	3040 Ci	3040 Ci

REACTOR STRONTIUM COMPOSITION

One of the uncertainties associated with the gravimetric method is the composition of reactor strontium. The composition changes significantly depending on the irradiation history of the fuel and the time since discharge. As an example, the composition of the fission product strontium in weapons-grade Hanford N Reactor fuel is shown in Figure 1 [4].

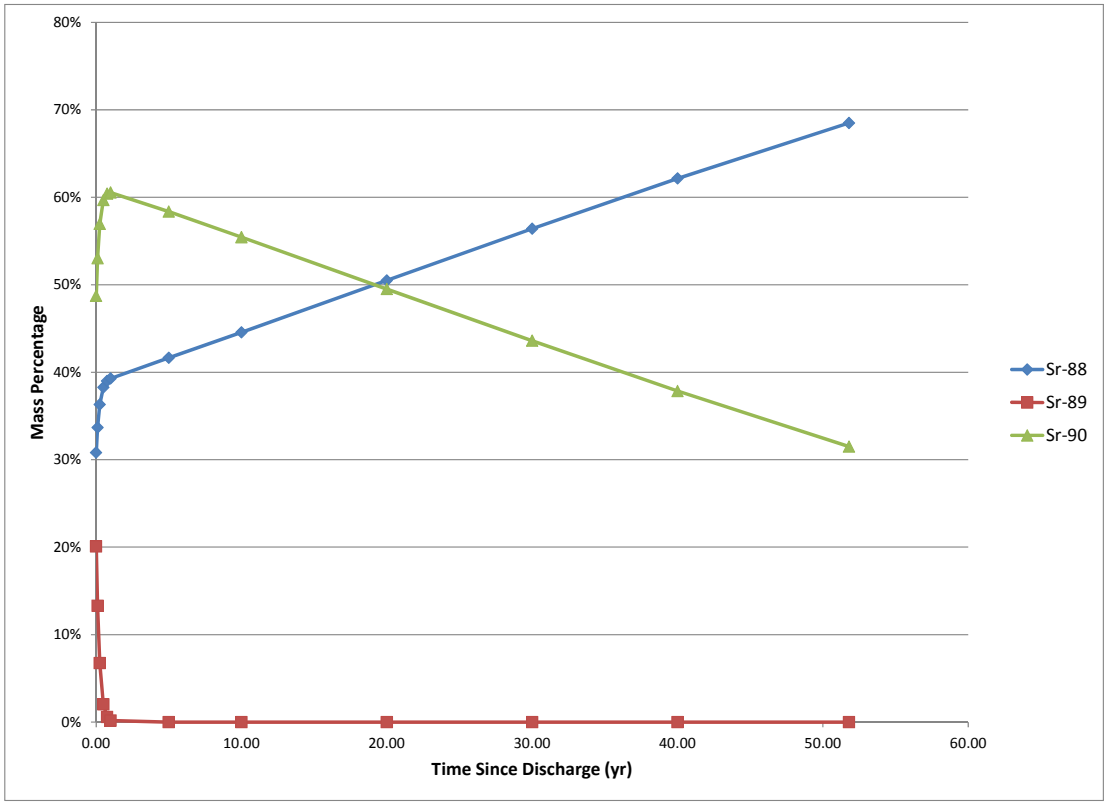

Figure 1. Reactor Strontium Composition

From Figure 1, the mass percentage of Sr-90 in reactor strontium changes from a peak of more than 60% shortly after discharge to just more than 30% after 50 years. The Sr-90 content predicted by gravimetric methods is linearly dependent on the Sr-90 mass fraction, and as a result, the estimated time since discharge.

FILTER DISPOSAL

The Sr-90 filters were retained for years as a potential source of additional Y-90 for medical isotope applications. Eventually it was determined that they were not needed, and they were declared waste. Waste disposal planning then began in earnest.

To support waste disposal efforts, it was necessary to determine the Sr-90 content in each filter. Although gravimetric estimates had been completed for all three filters, given the uncertainties associated with the gravimetric method an independent confirmation of the Sr-90 content was desired. Since it was not feasible to the sample the material in the filters for isotopic analysis, the only practical method for confirmation was dose rate to activity modeling.

DOSE RATE MEASUREMENTS

Dose rate measurements were first taken on the filters to support subsequent modeling. The dose rates were measured inside the hot cell, and in a location with as low a background as possible. The measured dose rates are shown in Table II.

Table II. Sr-90 Filter Dose Rate

Filter	On Contact	At 30 cm
Sr-01	250 R/hr	20 R/hr
Sr-02	1340 R/hr	180 R/hr
Sr-03	1050 R/hr	170 R/hr

The dose rates were measured using the Eberline RO-7 system [5], which features a hand-held instrument connected to a low-range, mid-range, or this case a high-range probe. Each probe consists of an air-filled ion chamber sized to provide the desired sensitivity, as well as a matched pre-amplifier capable of transmitting the signal through up to 500 feet of cable. Use of the RO-7 system allowed the probes to be placed in the hot cell while the instrument remained in the operating gallery.

MCNP MODELS

A series of MCNP [6] models were constructed for the filters in order to calculate the expected dose rate per unit Sr-90 activity. MCNP is a general-purpose, continuous-energy, generalized-geometry, time-dependent, coupled neutron, photon, and electron Monte Carlo transport code. In general, MCNP models consist of the following:

- A geometric representation of the problem
- The compositions of the materials in the problem
- The particle source term subject to transport

The same geometry and material compositions were used for all MCNP models. However, different particle source term options were compared to determine the optimum approach.

Geometric Model

Unfortunately, detailed construction drawings of the Sr-90 filters could not be located. Instead, the physical configuration of the filters was estimated from photographs of the filters in HLRF. Based on these photographs, the filters were modeled as 35.56 cm (14 in) tall, 10.16 cm (4 in) outer diameter, 0.635 cm (1/4 in) thick, and fabricated from Type 316L stainless steel. A sketch of the Sr-90 filter geometric model is shown in Figure 2.

The distribution of strontium carbonate within the filters was not known, and could only be estimated. Since the strontium carbonate in filters Sr-02 and Sr-03 was not retrieved, the radioactive source term for both filters was assumed uniformly distributed. The majority of the material in filter Sr-01 was retrieved for purification. Therefore, the radioactive source term for Sr-01 was assumed distributed in a 0.317 cm (1/8 in) thick layer attached to the walls of the filter.

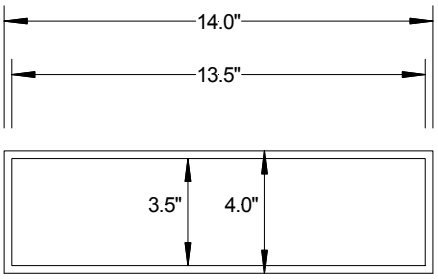

Figure 2. Sr-90 Filter Geometry

Materials

The filter model included air, 316L stainless steel, and strontium carbonate. The properties of air and 316L stainless steel were taken directly from PNNL-15870 [7], and were entered in terms of mass fractions. The chemical formula for strontium carbonate ($SrCO_3$) was used to enter the composition in terms of atom fractions.

The mass density of strontium carbonate was taken from the Material Safety Data Sheet (MSDS). The mass density for industrial grade strontium carbonate was stated to range from 0.3 to 0.7 g/mL. The wet precipitation method used to prepare the strontium carbonate in the filters was assumed to produce a material near the maximum end of the density range. Therefore a mass density of 0.7 was used in the MCNP model.

Source Term

For bremsstrahlung problems, MCNP may be operated in combined photon-electron (PE) or photon only (P) modes. The analyst has the option of providing the spectrum of beta particles and using MCNP to propagate both the electrons and the subsequently generated photons, or providing the photon spectrum from another code and using MNCP to propagate the photons alone.

The photon-electron mode has the benefit of being a stand-alone calculation, but the propagation of electrons in MCNP is a slow process and includes many approximations. The photon mode has the benefit of rapid execution, but requires an additional code to generate the photon spectrum. It was not clear at the outset which operating mode held the most promise. Therefore, models were constructed using both source term options, and the results were compared.

Mode PE

The beta spectra of the Sr-90/Y-90 decay chain are well established, and are available from numerous sources. Sources from different organizations and separated by many years were compared, and all gave essentially equivalent results. The spectrum from International Commission on Radiation Units and Measurements (ICRU) Report 56 [8] was selected for use.

The ICRU 56 spectra are presented as probability density functions, and are stated in units of beta particles/decay/(E/E_{max}), where E is the beta particle energy and E_{max} is the maximum beta particle energy. This representation has the benefit of unit independence, but must be converted to beta particles per decay per MeV for use with MCNP. Therefore each Sr-90 bin value was multiplied by 0.546 MeV,

and each Y-90 bin value was multiplied by 2.281 MeV. The resulting spectra were numerically integrated to ensure the area under the probability density function equalled unity.

The Sr-90 and Y-90 spectra were entered into the MCNP model as separate probability density functions, and each was sampled with a probability of 0.5. The WGT parameter was used to introduce the decay rate, which was set to 7.4E+10 to simulate 1 curie of Sr-90 and one curie of Y-90 in secular equilibrium.

Mode P

The photon spectrum for the mode P case was calculated using version 2.2 of the ORIGEN code [9]. An input file for 1 curie each of Sr-90 and Y-90 in secular equilibrium was constructed. The input file was then executed with the photon libraries for water and uranium oxide. Although the two spectra were similar in shape, the total photon rate was an order of magnitude larger for uranium oxide.

The photon rate from Sr-90 bremsstrahlung is linear with the average atomic number of the absorbing matrix. The average atomic numbers of water, strontium carbonate, and uranium oxide were calculated to facilitate interpolation. The average atomic number is given by:

$$Z_{eff} = \frac{\sum_i F_i Z_i}{\sum_i F_i} \qquad \text{(Eq. 1)}$$

In the above equation, F is a vector of the number of atoms of each element in the chemical formula, and Z is a vector of the atomic numbers of the corresponding elements. The average atomic numbers are given in Table III.

Table III. Average Atomic Numbers

Material	Z_{eff}
H_2O	3.333
$SrCO_3$	13.60
UO_2	36.00

The average atomic numbers were used to linearly interpolate between the water and uranium oxide spectra to yield the spectrum and photon rate for strontium carbonate. The photon spectra are shown in Table IV.

Table IV. Photon Spectra

Avg Energy (MeV)	H_2O (γ/sec)	$SrCO_3$ (γ/sec)	UO_2 (γ/sec)
1.00E-02	2.213E+09	9.387E+09	2.504E+10
2.50E-02	4.741E+08	1.985E+09	5.281E+09
3.75E-02	3.155E+08	1.304E+09	3.462E+09
5.75E-02	4.616E+08	1.889E+09	5.004E+09
8.50E-02	2.878E+08	1.152E+09	3.036E+09
1.25E-01	1.928E+08	7.573E+08	1.989E+09
2.25E-01	2.724E+08	1.033E+09	2.692E+09
3.75E-01	1.295E+08	4.628E+08	1.190E+09

Avg Energy (MeV)	H_2O (γ/sec)	$SrCO_3$ (γ/sec)	UO_2 (γ/sec)
5.75E-01	7.472E+07	2.477E+08	6.251E+08
8.50E-01	2.667E+07	7.954E+07	1.949E+08
1.25E+00	1.080E+07	2.751E+07	6.398E+07
1.75E+00	1.220E+06	2.406E+06	4.993E+06
2.25E+00	2.918E+02	3.733E+02	5.511E+02
2.75E+00	0.000E+00	0.000E+00	0.000E+00
3.50E+00	0.000E+00	0.000E+00	0.000E+00
5.00E+00	0.000E+00	0.000E+00	0.000E+00
7.00E+00	0.000E+00	0.000E+00	0.000E+00
9.50E+00	0.000E+00	0.000E+00	0.000E+00
Total	4.460E+09	1.833E+10	4.858E+10

The photon spectrum was entered into the MCNP model as a single histogram of discrete photon energies. The WGT parameter was set to a photon rate of 1.833E+10.

Tallies

Ring detector tallies were placed at 2.54 cm (1 in), 5.08 cm (2 in), and 30 cm from the surface of the filter. The 1 in and 2 in tallies were intended to simulate the on-contact dose rate measurement, and the 30 cm tally was intended to simulate the 30 cm dose rate measurement. Two tallies were used for the on-contact measurement to evaluate the sensitivity to measurement distance uncertainty. All tallies were converted to Roentgen per hour using the tally conversion factor and FM card method described in [10].

RESULTS

The MCNP input files described above were executed, and the results are listed in Table V.

Table V. MCNP Results

Case	Filter	Mode	Dose Rate (R/hr/Ci Sr-90)		
			1 in	2 in	30 cm
Case 1	Sr-01	PE	4.7838E-01	2.8932E-01	3.2466E-02
Case 2	Sr-02	PE	3.5574E-01	2.3125E-01	2.5692E-02
Case 3	Sr-03	PE	3.5574E-01	2.3125E-01	2.5692E-02
Case 4	Sr-01	P	5.8078E-01	3.9914E-01	5.4968E-02
Case 5	Sr-02	P	5.1702E-01	3.6522E-01	5.0351E-02
Case 6	Sr-03	P	5.1702E-01	3.6522E-01	5.0351E-02

The dose rate measurements in Table II may be divided by the MCNP factors in Table V to calculate the predicted Sr-90 activity in the filters. The results of these calculations are shown in Table VI.

Table VI. Sr-90 Activities

Case	Filter	Mode	Sr-90 Activity (Ci)		
			1 in	2 in	30 cm
Case 1	Sr-01	PE	522.6	864.1	616.0
Case 2	Sr-02	PE	3766.8	5794.6	7006.1
Case 3	Sr-03	PE	2951.6	4540.5	6616.8
Case 4	Sr-01	P	430.5	626.3	363.8
Case 5	Sr-02	P	2591.8	3669.0	3574.9
Case 6	Sr-03	P	2030.9	2875.0	3376.3

CONCLUSIONS

The Sr-90 activities in Table VI highlight a number of important conclusions:

- The use of on-contact dose rate measurements for waste characterization purposes should be discouraged for a number of reasons.
    - It is difficult to accurately reproduce instrument response in near-field conditions. The dose rate field can vary appreciably from one side of the detector to the other due to the steep gradient in the near field. This condition is in direct contrast to the uniform dose rate field used to calibrate the detectors.
    - The instrument response in the near field is very sensitive to the distribution of radioactive material within the object, and this distribution is often uncertain.
    - The instrument response is very sensitive to the distance from the detector to the object, and this distance is often not carefully controlled.
- The Mode P method coupled with the use of dose rate measurements at 30 cm agrees well with the gravimetric estimates made by the medical isotope project.
- The Mode PE method signficantly underestimates the dose rate produced by bremsstrahlung dominant radioactive materials, and should not be used. This phenomenon was attributed to the inability of the Mode PE method to incorporate photons from inner bremsstrahlung.

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