IMPACTS OF SHORT-LIVED RADIONUCLIDES ON 10CFR61 CLASSIFICATION AND DOSE-TO-CURIE METHODOLOGIES

Thomas P. Hillmer and James R. Hensch
Arizona Nuclear Power Project
Phoenix, AZ 85072

and

Alan D. Miller and Dean B. James
Advanced Process Technology
Milpitas, CA 95035

ABSTRACT

Procedures for dose-to-curie calculation and 10CFR61 classification must account for radwaste containing significant quantities of radionuclides with half-lives of weeks to months. The contributions from shorter-lived nuclides may be significant in waste from plants early in operational history. At the Palo Verde Nuclear Generating Station, the reactor coolant pumps contain antimony, and 60-day Sb-124 is the major nuclide in the radwaste. Computer calculations are presented that show how containers with a one-R/h dose rate but containing waste of different ages and with Co-60 and Sb-124 in varying ratios have very different total curie contents and compositions. Three approaches are described for accurately determining the curie compositions and 10CFR classification of such waste.

Short-lived radionuclides such as Co-58 and Sb-124 have long been known to cause difficulties in the determination of the isotopic compositions of the long-lived nuclides in materials from nuclear plants. The activities from the short-lived nuclides can mask to varying degrees the activities of the longer-lived nuclides. Also, dose-to-curie methodologies must account for the dose contributed by short-lived nuclides. This problem seldom arises if the analyses are conducted when the ages of the radioactive waste and the samples are all old relative to the age of the reactor coolant, and the shorter-lived nuclides have decayed to an extent that interference is no longer a problem. However, for plants early in their operational history, the contributions from short-lived activated corrosion product nuclides such as Cr-51, Mn-54, Fe-55, Co-58, Zn-65, and Sb-124 often comprise a significant fraction of the activity in the coolant and in the low-level waste even after several months of storage.

Unusual Antimony Levels at Palo Verde

The design at the Palo Verde Nuclear Generating Station (PVNGS) has included reactor coolant pump seals that contain antimony. Corrosion products containing this antimony are transported via the primary coolant to the fuel where they are deposited and activated. The corrosion products are later released to the primary coolant and are deposited on the out-of-core surfaces or removed by the reactor coolant purification system. The activated isotopes of antimony then contribute significantly to the radiation dose and curie inventory of the radwaste system.

The natural antimony is activated by the neutron flux to form two radioactive nuclides. Sb-122 is created from the natural Sb-121 (57.3%), but it has only a 2.8-day half-life, so it is not a significant source of activity for low-level waste. Sb-124 is created from the natural Sb-123 (42.7%). However, Sb-124 has a 60-day half-life, and when it is released to the coolant, it contributes significantly to both occupational exposure and radwaste disposal costs. Nuclear plants without the large source of antimony do not have the extra burden from the activated antimony isotopes. The unusually high levels of Sb-124 in the Palo Verde radwaste provides an example of how the shorter-lived isotopes can impact the conventional radwaste procedures.

Impact of Sb-124 on Dose-to-Curie Methods

Antimony-124 has a sufficiently long half life, and a sufficiently energetic and abundant gamma ray emission to impact significantly the dose-to-curie determination methodology for radwaste. This can be shown by calculating the curie contents and curie compositions of one R/h container of different ages. For a container dose rate of one R/h for all ages of the waste, and for three different initial ratios of Sb-124 to Co-60, the calculated curie contents of such containers are shown in Figs. 1 and 2.

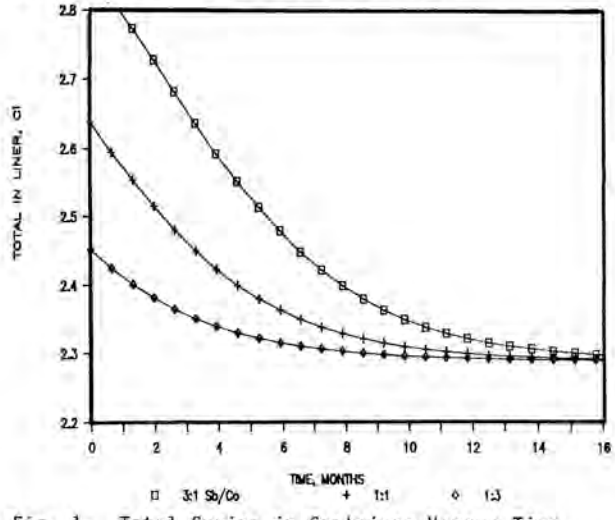

Fig. 1. Total Curies in Container Versus Time

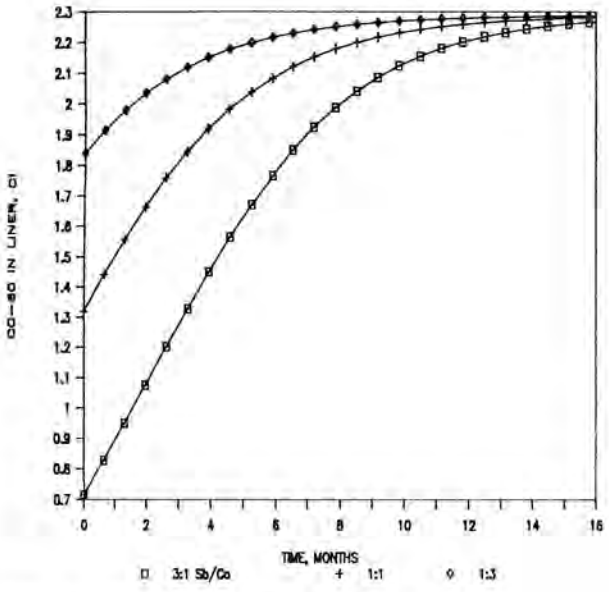

Fig. 2. Co-60 in Container Versus Time

Figure 1 shows how the total curie content of a 1-R/h container varies with the age of the radwaste it contains for the three different initial Sb-124/Co-60 ratios. The Sb-124 decays in accordance with its shorter half life. In the time period that radwaste is usually stored, the decay of Co-60 is negligible. As the storage (decay time) increases, the nuclide distribution changes with the Co-60 completely dominating after a long time. As can be seen from the trend in the curves, a container containing about 2.3 Ci of Co-60 and negligible Sb-124 would have a dose rate of one R/h and an unchanging total curie content.

Figure 2 shows the varying Co-60 curie content of one-R/h container for the three starting ratios of Sb-124 to Co-60. The aged radionuclide distribution must be known before an accurate dose-to-curie determination can be made. Also, since Co-60 is the key radionuclide for determining the concentrations of several non-gamma-emmitting isotopes, the scaling factor methodology of 10CFR61 is directly impacted by any uncertainties in the Co-60 content caused by the decay of such shorter-lived isotopes such as Sb-124.

Dose-to-curie calculations must not be based on the classic assumptions that the only significant gamma emitters in waste are the long-lived isotopes like Co-60. To be defensible, dose-to-curie calculations will have to be based on measured, decay-corrected fractional isotopic compositions that include all the gamma emitters including those that are short-lived and could make any significant contribution to the measured dose rate. Conservatism in these calculations can lead to unnecessary disposal costs.

Several methods for accounting for the effects of these nuclides on radwaste operations, radiochemical analyses, and waste disposal are being implemented at PVNGS. The contributions of Sb-124 and other short-lived radionuclides to the dose-to-curie methodology employed for the disposal of dry active waste and cartridge filters require that either the waste components be tracked individually or that gamma-scan measurements of the contents of each container be made at or near the time of waste shipment to determine the relative radionuclide distributions.

Radwaste Management With Short-Lived Contributors

Several solutions to the problem of tracking the levels of short-lived isotopes and the changing isotopic fractions are possible:

1) the waste could be characterized by (a) taking periodic representative samples and decay-correcting the non-gamma scaling to the time of generation, and (b) decay correcting the calculated nuclide composition to the time of shipment for the dose-to-curie determination;

2) the waste could be sampled and analyzed at the time of shipment;

3) a direct nondestructive gamma-assay technique could be employed to measure accurately the fractional curie composition of the gamma-emitting nuclides in each container at time of shipment.

Table I lists the advantages and disadvantages of these three options.

A scheme for approach 1 is illustrated in Fig. 3. for filter media. While the calculations involved are lengthy, individual tracking of waste components is adequate for the determination of the activity distribution of a multi-component container if provisions are included in procedures for recording the waste generation date and dose-rate contribution of each component of the waste. The process could be computerized using a spreadsheet-type program. A

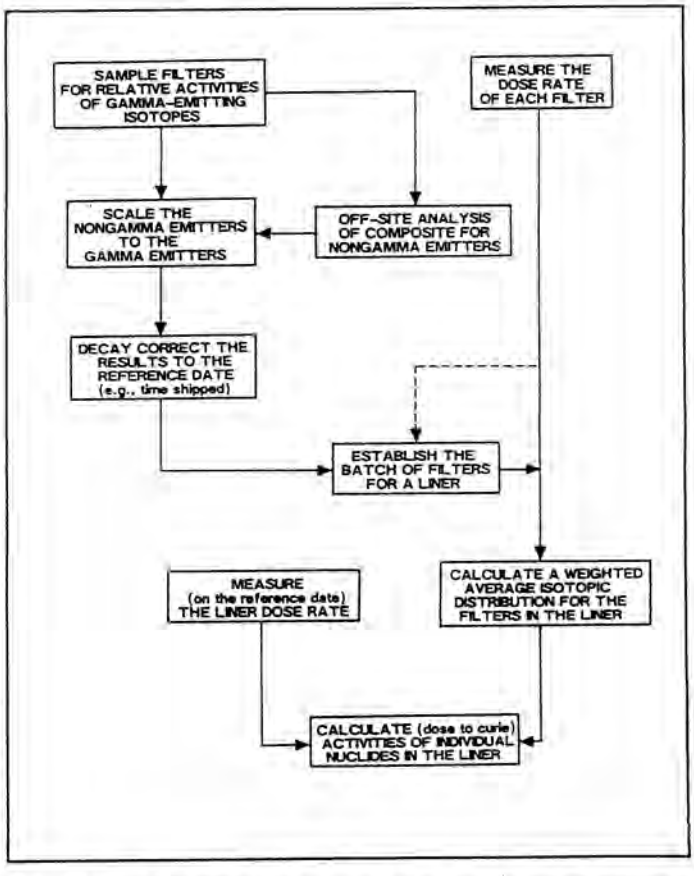

Fig. 3. Determination of the Total Activity of Filter Media in Waste Shipments

weighted isotopic distribution for a liner would be based on the dose rates of the individual filters. Care would have to be taken that all waste is of approximately the same or of known age since a correction must be applied for the radioactive decay of the waste ralative to the samples used for the scaling factor determination. The decay corrections can only be applied reliably if the ages of each container of waste are known and can be used when the decay corrections are made on the date of shipment. The radwaste procedures would have to contain provisions for documenting the waste generation dates and for retaining these data for use in the decay corrections that must accompany the dose-to-curie calculation.

Approach 2 could cause delays, may not be operationally feasible for some waste forms, and the exposure penalties may be large. But, for streams that are easily and reliably sampled, sampling at or near the time of shipment might be considered. Sampling of waste forms such as filter cartridges at the time of shipment probably would not be feasible from either ALARA considerations orfrom a representativeness standpoint. One of the other techniques must be employed for these types of waste.

At the PVNGS nondestructive gamma scanning of a representative selection of container types and waste forms will be tested. This will evaluate the feasibility of approach 3, and will provide a cross-check "calibration" of the sample/decay-correction approach. The feasibility study will determine whether time and dose-commitment considerations will justify the capital cost. The results may show that certain waste forms or container types might be more easily assayed directly rather than using a system that must compile and track a large amount of data. The gamma scan technique could be used to determine the relative abundances of the gamma emitting iosopes or, with proper calibration, the total contents of the waste.

TABLE I

Radwaste Measurement Options

Option	Advantages	Disadvantages
1. Sample at the time of shipment.	- Avoids decay and dose-to-curie calculations.	- Gives ALARA problems from the higher exposures. - Very difficult to get a representative sample.
2. Track individual waste-components from periodically measured distributions and then apply decay corrections and dose-to-curie calculations.	- Accounts for varying quantities and distributions in waste. - Sampling errors are minimized by incorporating multiple samples. - Can be used to forecast the curie contents prior to placing waste in container.	- Requires periodic sampling and tracking of data for each waste form, for each waste source and its distribution of activity and relative quantity of activity.
3. Nondestructive gamma-scan measurement at the time of shipment.	- Potentially most accurate. - Avoids lengthy record keeping and tracking of various waste components. - Avoids decay calculations. - Avoids dose-to-curie calculations (if used quantitatively). - Eliminates waste form-sampling problems for gamma isotopic distribution. - Eliminates radiation exposures from the taking of individual samples.	- Requires purchase, calibration, and operation of relatively complicated and expensive equipment. - Requires preparation of representative standards with known contents for each waste form and container type (if used quantitatively). - Requires careful design of the measurements set-up to minimize radiation exposure during measurement.