

## CURRENT STATUS OF LIQUID SCINTILLATION AT WESTINGHOUSE HANFORD COMPANY

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**ABSTRACT.** We present here an overview of the current applications of liquid scintillation counting (LSC) performed at Westinghouse Hanford Company. Our goal is to ascertain, with the greatest reliability possible, the activity contained in a sample. We measure samples for unconditional release criteria. We also test experimental processes to determine the effectiveness of removing transuranics from liquids by precounting and postcounting spiked solutions. We use a variety of methods to count soils, oils, mixed waste, water samples and other organic and inorganic materials. Some methods require a wet chemistry separation, whereas other methods may be mixed directly with a cocktail and counted. Using LSC, Westinghouse Hanford Company can detect activities below current federal regulatory thresholds and, therefore, release liquids unconditionally.

### INTRODUCTION

The Hanford Site covers approximately 1500 km<sup>2</sup> in southeastern Washington State. It was established in the early 1940s for full-scale plutonium production. After nearly 50 years of operation, the mission of the site has become environmental restoration, cleanup and research. Westinghouse Hanford Company (Westinghouse Hanford) is the U.S. Department of Energy's (DOE) operations and engineering contractor at the Hanford Site.

Currently, five facilities at Westinghouse Hanford use liquid scintillation counting (LSC) for measuring both alpha and beta radiations. Isotopes presently measured include <sup>241</sup>Am, <sup>14</sup>C, <sup>3</sup>H, <sup>125</sup>I, <sup>237</sup>Np, <sup>147</sup>Pm, <sup>238</sup>Pu, <sup>240</sup>Pu, <sup>241</sup>Pu, <sup>75</sup>Se, <sup>79</sup>Se, <sup>90</sup>Sr, <sup>99</sup>Tc and <sup>232</sup>U. We also measure total activity, gross  $\alpha$  and gross  $\beta$ . These radionuclides come from sources such as liquid effluents from facility processes, liquid and solid waste, and environmental sampling. The samples may be organic or inorganic (e.g., water, filter paper, oils and air effluents). We are also developing methods to measure <sup>137</sup>Cs, <sup>63</sup>Ni and <sup>106</sup>Ru. Table 1 summarizes LSC data for these radionuclides.

Our objective is to use LSC as a resource for releasing materials to offsite locations and for measuring radioactivity with the greatest possible accuracy. Present applications include sample scanning, characterization for cleanup activities and personnel protection. A special application is determining Pu removal *via* separation processes. For this application, we measure a sample using LS before and after the extraction process to determine the efficiency of removal of Pu and other transuranics (TRU). We will pursue other applications such as photon/electron rejecting alpha liquid scintillation (PERALS®) and pulse-shape discrimination and separation. Optimal relations between vials, cocktails and sample volumes will be determined. Finally, screening for radiological postings, analysis of conditions, planning, release criteria and sampling will become routine and accepted.

### METHODS

Because we use different types of LS counters, the region of interest (ROI) is reported in keV for Packard LS counters and in channels for Beckman LS counters (Table 1). Also, quench factors are reported in transformed spectral index of external standard (tSIE) for Packard LS counters and in H numbers for Beckman LS counters.

TABLE 1. Summary of Liquid Scintillation Counting at Westinghouse Hanford Company

	ROI	FM = E <sup>2</sup> /B	Quench	Count time (min)
<i>Total Activity</i>				
Oils	0–200 keV	338	tSIE~160	60
	200–600 keV	2500		
	0–1000 channels	135	H no. 160->500	10
Water	0–300 keV	347	tSIE~265	30
	300–600 keV	3008		
Soil/mixed solids	0–1000 channels	135	H no. 100–200	10
Hazardous waste	5–1000 channels	26–111	H no. 85–465	5
<i>Alpha Counting</i>	70–500 keV	~533	Variable tSIE	5–500
<i>Beta Counting</i>				
<sup>14</sup> C	0–700 keV	135–205	H no. 0–250	30
Filter paper	2–18.6 keV	335	--	4
Liquid effluents	0–400 channels	59	H no. ~110	30
<sup>123</sup> I	0–600 channels	23–184	H no. 70–225	10
<sup>63</sup> Ni	0–700 channels	18–98	H no. 50–300	10
<sup>147</sup> Pm	0–700 channels	142–184	H no. 60–225	10
<sup>75</sup> Se	0–700 channels	6–61	H no. 80–340	5
<sup>79</sup> Se	0–700 channels	142–201	H no. 0–250	30
<sup>99</sup> Tc	150–800 channels	131–146	H no. 70–210	10

Gross alpha and beta counting and measuring total activity are done routinely to screen liquids for unconditional release from the Hanford Site. To release a substance, its total activity must be <7.4 Bq g<sup>-1</sup>, and the total  $\alpha$  activity must be <2.22 Bq g<sup>-1</sup>. Released substances include various oils, water, soils, mixed solids and other hazardous waste.

Oils include motor oil, hydraulic oil, oil from waste pads and paints. No wet chemistry or mechanical separations for oils are performed. Typically, we fill 20-ml low-potassium borosilicate glass LS vials with 15 ml of Opti-Fluor® (Packard Instrument Co.) and add an oil sample of up to 5 ml or 1 g. We cap the sample with a Teflon-coated cap, shake it, and allow the sample to dark-adapt for a minimum of 12 h. We vary the amount of sample because dark oils tend to have large quenching. The smallest amount of a dark oil used has been 0.33 ml. Light oil samples are generally 5 ml or 1 g.

After the samples have dark-adapted, we count them for either 60 min each on a Packard LS counter, or for 10 min each on a Beckman LS counter. For a Packard LS instrument, we have set the ROIs at 0–200, 200–600 and 600–2000 keV. This yields backgrounds of 24, 4 and 5 counts per minute (cpm), respectively. The  $\beta$  efficiency, in the first ROI, is ~90%. The  $\alpha$  efficiency, in the second ROI, is near 100%. The typical quench factor is a tSIE of 157. An open window of 0–1000 channels is used for a Beckman LS instrument. The background is ~60 cpm for the open window and the efficiency is ~90% using a <sup>99</sup>Tc standard, but depending upon the quench, the efficiency may drop to 0%. The background is slightly less efficient than using <sup>90</sup>Sr as the  $\beta$  standard.

We prepare water samples with 15 ml of Ultima Gold™ (Packard Instrument Co.) and 5 ml of sample in 20-ml borosilicate low-potassium glass LS vials. We count these samples for 30 min without waiting for dark-adaptation. Using a Packard LS counter, the ROIs are 0–300, 300–600 and 600–2000 keV. The background in each of the ROIs is 26, 3 and 6 cpm, respectively. Both the

first and second ( $\beta$  and  $\alpha$  ROIs, respectively) ROIs have an efficiency of  $\sim 95\%$  and the tSIE is  $\sim 265$ . Water that has an extreme pH, either basic or acidic, tends to turn the sample milky, increasing the quenching.

We prepare soil and mixed solid samples by directly mixing 1 g of sample with 15 ml of Opti-Fluor® in a 20-ml borosilicate low-potassium glass LS vial. After waiting 12 h for dark-adaption, we count the samples for 10 min each, using a Beckman LS counter with an open window (0–1000 channels). The background is  $\sim 60$  cpm and the efficiency is near 90%, using a mixed  $^{99}\text{Tc}$  standard. In the future, we plan to add 1 ml of water to the 1 g of sample before adding it to the cocktail to monitor potential leaching that would increase our ability to measure the total activity in soils and mixed solids.

For hazardous waste samples, we mix 19 ml of Insta-Gel® XF with either 1 g or 1 ml of sample in a 22-ml low-potassium borosilicate glass LS vial. After 3 h of dark-adaptation, the samples are counted for 5 min, during which we are able to determine if a sample is below the federal regulatory threshold. The background is  $\sim 90$  cpm in the ROI of 5–1000 channels. The efficiency ranges between 100% and 49%, and the H number ranges between 84 and 464, respectively. We determine efficiency by using a mixed  $^{60}\text{Co}$  and  $^{241}\text{Am}$  standard. We can readily detect  $^{90}\text{Sr}$  and screen hazardous waste with this method.

Currently, we use  $\alpha$  LS counting to test TRU removal processes. We perform wet chemistry separations to isolate  $^{241}\text{Am}$ ,  $^{237}\text{Np}$ ,  $^{239}\text{Pu}$ ,  $^{240}\text{Pu}$ ,  $^{241}\text{Pu}$  and  $^{232}\text{U}$ , and use  $^{238}\text{Pu}$  as a tracer. We verify the removal of actinides such as Pu and Am by counting spiked solutions (e.g., waste water and tank sludge) before and after a process is applied. Waste water, tank sludge and other hazardous waste are processed to reduce the volume of TRU waste to below regulatory thresholds. The following limits apply to TRU:  $<3.7$  kBq  $\text{g}^{-1}$  is considered low-level waste;  $>3.7$  kBq  $\text{g}^{-1}$  is considered high-level waste or TRU. In theory, we should be able to show a reduction of waste by 95%, leaving only 5% to be treated as high-level waste and vitrified. We are now considering using LSC to test processes for removing  $^{137}\text{Cs}$ ,  $^{106}\text{Ru}$ , and  $^{90}\text{Sr}$  (although these isotopes are not  $\alpha$  emitters).

Spiked  $\alpha$  samples are prepared using 15–16 ml of Ultima Gold™ and 1 ml of sample in a 20-ml LS glass vial with an aluminum-lined cap. The activity of a spike ranges from 50 kBq to 17 mBq. Three ROIs have been set up: 0–70, 70–500 and 0–1000 keV. The first ROI is the  $\beta$  window, the second ROI is the  $\alpha$  window, and the last ROI is a gross  $\alpha$  and  $\beta$  window.  $^{241}\text{Pu}$  is the most active of the Pu isotopes and emits both  $\beta$  and  $\alpha$  particles (Fig. 1). The  $\beta$  efficiency is between 10 and 40%, depending upon the quench.  $^{241}\text{Am}$  is a pure  $\alpha$  emitter, distinguished from Pu by yielding counts only in the  $\alpha$  ROI. The counting efficiency for the  $\alpha$  particles is near 100%. We count prepared samples without waiting for dark-adaption. Depending on the activity, we may count a sample from 5–500 min. The  $\alpha$  background is  $\sim 18$  cpm, whereas the  $\beta$  background is  $\sim 80$  cpm.

Single-shell tank, environmental and record samples provide the basis for  $^{14}\text{C}$  LS counting. We prepare samples using 22-ml low-potassium borosilicate glass LS vials and filling them with 5 ml of Carbo-Sorb® (Packard Instrument Co.), an amine for extracting  $\text{CO}_2$ , and 15 ml of Insta-Gel® XF. We cap samples with a Teflon-coated cap and allow the samples to dark-adapt for 3 h. We count the samples for 30 min each, with ROI of 0–700 channels. The efficiency ranges between 96 and 78% according to the quench. The H number is between 0 and 250, respectively. In the ROI, the background is  $\sim 45$  cpm.

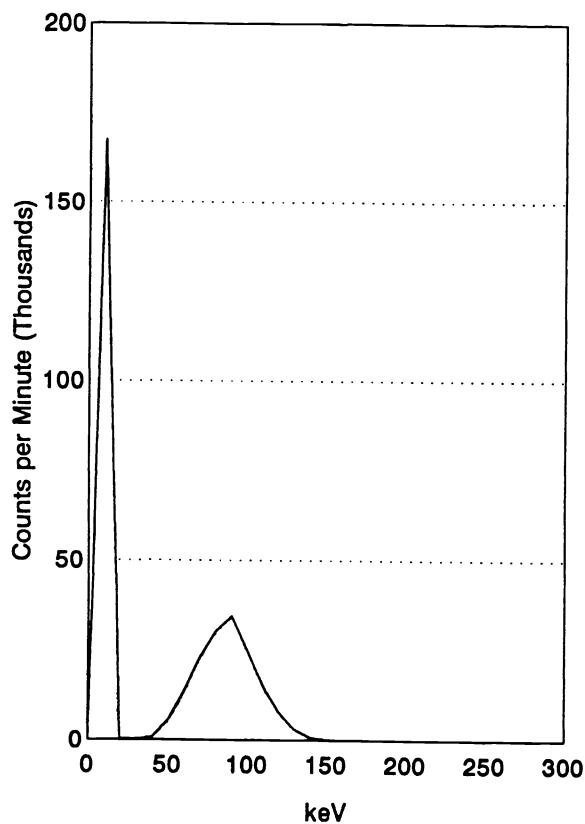


Fig. 1. Typical spectral output of  $^{241}\text{Pu}$

$^3\text{H}$  samples come from survey smears, liquid effluents and environmental sampling. We take 100-cm<sup>2</sup> smear samples on 42.5-mm Whatman Grade No. 5™ filter paper and the smear samples are counted for offsite release purposes. Smear samples must be <17 Bq total activity to be released unconditionally. Liquid effluents and environmental samples that are >7.4 Bq g<sup>-1</sup> go through additional analysis.

For smear samples, we push the filter paper to the bottom of a 20-ml glass LS vial. We then add 10 ml of Ultima Gold™ and cap the sample with a Teflon-coated cap. The filter paper is transparent to ultraviolet light and partially dissolves in the cocktail. We count the samples for 4 min after the samples have dark-adapted for a minimum of 1 h. In the ROI, 2–18.6 keV, the background is ~13 cpm; efficiency is ~66%.

We prepare samples from liquid effluents and the environment by using 1–5 ml of sample and filling the remainder of the 22-ml low-potassium borosilicate glass LS vial with Insta-Gel® XF. We seal the samples with a Teflon-coated cap and allow the samples to dark-adapt for 3 h. We count samples for 30 min each, with ROI of 0–400 channels. The efficiency ranges between 4 and 65% with H number ranging from 330 to 10, respectively. The average efficiency is 40% with an H number of 110; background is ~27 cpm. The 1-ml samples mixed with Insta-Gel® XF remain a liquid, whereas 5-ml samples mixed with Insta-Gel® XF become a gel.

We are currently developing a method to determine the presence of  $^3\text{H}$  using 1 ml of sample and 6 ml of cocktail in a 7-ml LS vial, so as to reduce the volume of waste. No data are available at this time.

Occasionally, standards laboratories request the verification of a tracer, such as  $^{125}\text{I}$ . We prepare samples by mixing 1 ml of sample with 19 ml of Insta-Gel® XF in a 22-ml low-potassium borosilicate glass LS vial. A Teflon-coated cap is used to seal the samples. After the samples dark-adapt for 3 h, they are counted for 10 min each with ROI of 0–600 channels. The efficiency ranges from 91 to 32% with the H number ranging from 73 to 226, respectively. The background in the ROI is ~45 cpm.

Currently, we are developing a  $^{63}\text{Ni}$  procedure, which will be used for radioanalytical standards verification. We have not yet established the aliquot and cocktail volumes, but, we believe a 1-ml aliquot of  $^{63}\text{Ni}$  solution and 19 ml of Insta-Gel® XF will be added to a 22-ml low-potassium borosilicate glass LS vial. We plan to count samples for 10 min each after they are allowed to dark-adapt for a minimum of 3 h. In the expected ROI of 0–700 channels, the background is ~50 cpm. The measured efficiency ranges between 30 and 70%, as does the H number from 50 to 300, respectively. Figure 2 shows the quench curve spectral output of  $^{63}\text{Ni}$ .

Because  $^{147}\text{Pm}$  has a short half-life, 2.6 yr, we do not get many requests for its analysis. However, we receive samples from one of the standards laboratories, the waste tank facilities and from plume stack effluents. Using 22-ml low-potassium borosilicate glass LS vials, we prepare samples by mixing 5 ml of 1.23 M nitrate  $^{147}\text{Pm}$  solution with 15 ml of Insta-Gel® XF. This is a high-acid solution and may cause significant oxidation and quenching. We count samples for 10 min each after they dark-adapt for 3 h. The ROI is 0–700 channels. The efficiency, which ranges from 91 to 80%, does not vary much with H number, which ranges from 63 to 225. Background is ~45 cpm.

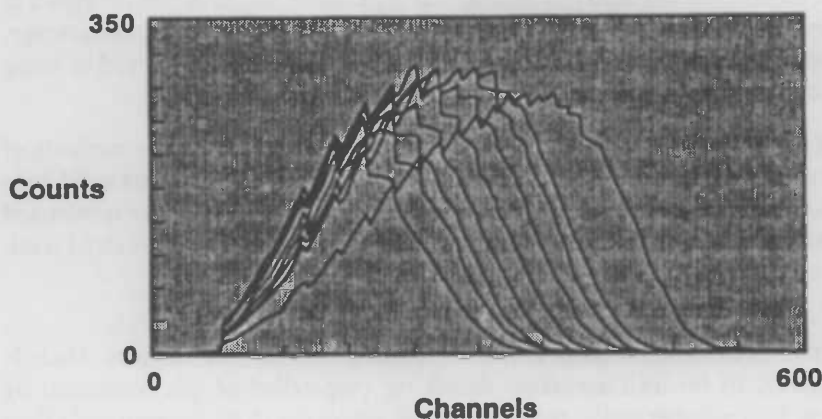


Fig. 2. Quench curve spectral output of  $^{63}\text{Ni}$

We first leach samples from waste tanks, which require  $^{75}\text{Se}$  determination. The  $^{75}\text{Se}$  and  $^{125}\text{I}$  sampling techniques are similar. We prepare  $^{75}\text{Se}$  samples using 22-ml low-potassium borosilicate glass LS vials filled with 1 ml of sample and 19 ml of Insta-Gel® XF. Teflon-coated caps are used to seal the samples. We count samples for 5 min each after they dark-adapt for 3 h. The ROI is between 0 and 700 channels, and the background is ~100 cpm. The efficiency ranges from 78 to 25% and varies with the H number, between 80 and 340, respectively. To measure  $^{79}\text{Se}$ , we first adsorb it onto a glass fiber filter, which is then leached in a 1-ml aqueous solution. Samples are prepared using 22-ml low-potassium borosilicate glass LS vials with Teflon-coated caps. The 1-ml sample is mixed with 19 ml of Insta-Gel® XF. After the samples dark-adapt for 3 h, they are counted for 30 min each. The ROI is between 0 and 700 channels and the background is ~45 cpm. The efficiency ranges between 95 and 80% and varies with the H number from 0 to 250.

$^{99}\text{Tc}$  samples come from liquid effluents and waste tanks.  $^{99}\text{Tc}$  is also used in tracer studies. We prepare samples using 22-ml low-potassium borosilicate glass LS vials with Teflon-coated caps. A 4-ml aliquot of sample is mixed with 16 ml of Insta-Gel® XF. Each sample is counted for 10 min after an initial dark-adaption period of 3 h. The ROI is between 150 and 800 channels, and the background is ~63 cpm. The efficiency, which ranges between 96 and 91%, varies with the H number, which is between 73 and 210.

We recently received a PERALS® spectrometer, which incorporates pulse-shape analysis techniques with LSC to eliminate the  $\alpha$ - $\beta$  discrimination problem. The PERALS® method distinguishes characteristic light-pulse shapes, "signatures", produced in the scintillator by the respective type of ionizing radiation. The duration of the photon pulse is indicative of the specific ionization. Alpha pulses are 30 nsec longer than  $\beta$  pulses. Except for oxygen, the chemical, color and dilution quenchers reveal that moderate quenching does not adversely affect pulse-shape analysis. Further, only the  $\alpha$  light pulse is affected by oxygen (McKlveen & Johnson 1975).

We will use PERALS® to study environmental samples and evaluate TRU removal. The former includes the analysis of radionuclides in washed soils and waste tank sludge using extractive scintillators. Using extractive scintillators for specific nuclides is an advantage, as is the reduction in scintillation cocktail.

## CONCLUSION

We have discussed the measurement of a variety of radionuclides. Some methods are quick and simple, whereas others require wet chemistry or mechanical separation. We have not attempted to discuss all of the chemical or mechanical separations, because of space constraints. On average, we process ~2500 samples per month. By using new instruments, such as PERALS®, and by using small sample vials, such as for  $^3\text{H}$ , we are reducing the waste produced by LS.

An important note regarding LSC is that it may be used in conjunction with other methods of radiation detection, which include gas flow proportional counting,  $\gamma$  spectrometry and solid-state surface barrier detection. Using a variety of methods increases the reliability of our results and improves our ability to determine whether a sample is above or below a regulatory threshold level.

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## REFERENCE

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