Non-destructive In situ Measurement of Radiological Distributions in Hanford Site Waste Tanks

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Non-destructive *In situ* Measurement of Radiological Distributions in Hanford Site Waste Tanks[1]

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Abstract

Measurement of radiological materials in defense nuclear waste stored in underground tanks at the Hanford Site is being used to indicate material distributions. Both safety assessment and future processing challenges are dependent on knowledge of the distribution, kinds, and quantities of various key components. Data from CdTe and neutron detector measurements are shown and correlated with physical sampling and laboratory results. The multiple assay approach is shown to increase the confidence about the material distributions. As a result, costs of physical sampling and destructive analyses can be controlled while not severely limiting the uncertainty of results.

I. INTRODUCTION

A. Application

Remote *in situ* cadmium telluride (CdTe) and similar CdZnTe detector gamma-ray spectroscopy is useful for radionuclide characterization within high dose rate environments where access space is limited. The small size, ruggedness, room temperature operation, moderate resolution, and high atomic number of CdTe and CdZnTe detectors allow for high energy gamma-ray spectroscopy in environments where conventional germanium and sodium iodide spectroscopy would be difficult.

In situ CdTe spectroscopy has been used in the characterization of high level nuclear waste tanks (HLWTs) in which the U.S. Department of Energy stores a large portion of the nation's high-level radioactive waste. A general configuration of an HLWT is shown in Fig 1. HLWTs contain up to one million gallons of highly radioactive chemical wastes from nuclear fuel reprocessing at the Hanford Site. *In situ* CdTe spectroscopy has been used to measure gamma-ray-emitting radionuclide content and distribution within HLWTs. This information is used to aid in tank characterization and is being developed for a process monitor of the effectiveness of waste retrieval.

[1]Work performed for the U.S. Department of Energy under Contract DE-AC06-87RL10930.

CdZnTe detectors are used for analytical screening of HLWT core samples within a Hanford Site remote access hot cell. The data generated yield information about the homogeneity of the sample. The information is used to assist the laboratories in sub-sampling the core effectively for further analysis. Data are also used in shipping sub-samples to offsite laboratories for analysis.

Additionally, CdTe and CdZnTe detectors have been used for general non-destructive assay in highly radioactive

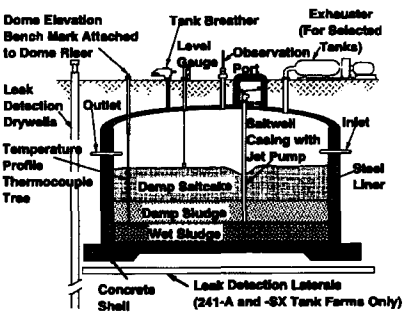

Fig. 1 Typical Hanford Site high-level nuclear waste tank

environments where access is limited. *In situ* gamma ray spectroscopy has been performed for the characterization of various Hanford Site items: a sand filter at the 105-KE spent fuel storage basin; fuel element spacers at N-reactor; and a storage tank at a pilot plant.

B. Detector Configuration

Normally a remote probe contains the detector, shielding, preamplifier, and when needed, an auxiliary cooling system. All other signal processing and computer equipment is located at a distance. Measurements are often performed through access pipes between 6.4 and 15 cm (2.5 and 6 in) in diameter. Detectors are often placed in a collimating shield to limit the count-rate and define the field of view [1,2,3].

CdTe and CdZnTe detectors have the undesirable characteristic of charge trapping within the detector elements, which normally results in severe spectral degradation at high gamma-ray energies. This effect has been partially overcome by a technique called pulse risetime compensation (PRC). PRC is accomplished through a unique pulse processing system configured from commercially available nuclear instrument modules. The basic configuration is shown in Fig. 2.

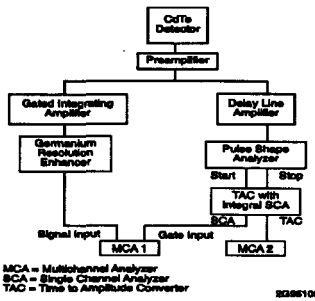

Fig. 2 Block diagram of a pulse shape processing system

The PRC technique compensates for charge trapping by adjusting the amplitude of the detector pulse by an amount dependent on the risetime [1,3,4-7]. A small percentage of the pulses with extremely long risetimes originating from events with high degrees of charge trapping are generally rejected using pulse shape discrimination (PSD).

Typical signal processing electronics (Fig. 2) are composed of five nuclear instrument modules between the preamplifier and the multichannel analyzer (MCA). The typical system has two major sections for PRC and a PSD. The PRC section has a spectroscopy amplifier, followed by an EG&G Ortec model 675 germanium resolution enhancer (GRE). The GRE adjusts the pulse amplitude as a function of charge collection time. Output from the GRE is to a standard MCA that stores the spectrum. The PSD electronics gate the MCA to reject approximately 10% of the total pulses that have risetimes too long for effective compensation. PSD electronics consist of a delay line amplifier, a pulse shape analyzer/timing-single channel analyzer, and a time-to-amplitude converter/single channel analyzer (TAC/SCA).

C. Gamma Spectroscopy

Data reduction is generally based on the net photopeak area. Due to the intense and predominant signal from ^{137}Cs gammas and ^{90}SrY bremstrahlung in the HLWT waste, minor component photopeaks of interest have limited counts and are generally on top of a large and steeply sloped background. The initial application of various commercial analysis packages was unsuccessful. Typical polynomial, straight line, and stepped background fits were inadequate, and produced erratic fits of the background. A suitable method for determining the background underneath the photopeak is to take the logarithm of the background channel numbers, then in each peak region fit a third order least squares curve to the background counts. APTEC Nuclear Inc. has included this option in their commercial gamma-ray spectroscopy software for fitting the background to photopeaks.

D. Shielding, Collimation and Cooling

A typical shielding and collimation arrangement is depicted in Fig. 3. The primary needs for HLWT measurements are small diameter, significant shielding, vertical spatial discrimination, and when needed, moderate cooling. The spatial collimator and shielding are adjusted to meet specific needs by changing the thickness of the plastic spacer and/or by changing shielding thicknesses. Shielding is generally designed to keep the count-rate to less than 100k counts per second and must address general field strengths up to 11 Gy/hr.

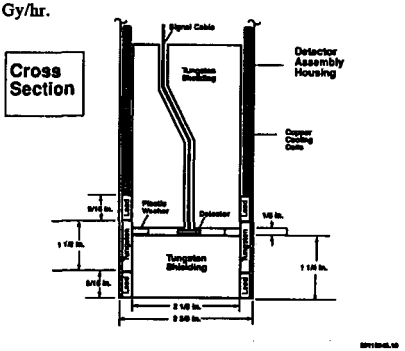

Fig. 3 CdTe detector assembly for HLWT well logging

Radioactive decay in the waste results in elevated temperatures up to 60 °C (140 °F) within some waste tanks. While the 2x2x2-mm CdTe detector selected for this application operates well at room temperature, spectral degradation at such elevated temperatures occurs and is unacceptable. When necessary the detector is cooled by circulating ice water through cooling coils. Freezing the heavy collimation and shielding apparatus and then using it within a stainless steel vacuum dewar is also successful. Operational times of several hours have occurred for the latter method.

E. CdTe Versus CdZnTe

CdZnTe and CdTe detectors are very similar and there is not a large advantage of using either detector material for the noted applications. CdZnTe has a small advantage over CdTe in that it can be operated at a higher bias voltage and the degree of trapping is somewhat less. However, among the detectors used by Westinghouse Hanford Company (WHC), a CdTe detector yielded the best resolution.

CdTe detectors have proven to be very rugged. One particular detector is still in use after 6 years. It has been

subject to the abuse of numerous drops, over-biasing, temperatures up to 65 °C (150 °F) while under-bias, saturating gamma-ray fluxes, and mechanical stresses related to tightly fitting shields. No detectors of either material have failed at WHC, except for one CdTe detector that was subjected to intentionally severe over-biasing. CdZnTe detectors have not yet stood up to the tests of time and abuse at the Hanford Site. Experience has shown the detector-to-detector variations are large enough that detectors must be individually selected for each application. The principle detector used for most of the applications has been a 2x2x2-mm CdTe detector.

F. CdTe Versus Traditional Detectors

The resolution of CdTe and CdZnTe detectors is significantly better than the standard sodium iodide scintillators, but not as good as conventional germanium semiconductor detectors. Scintillation detectors yield spectra with photopeak resolution that is not well suited for most applications. While the higher resolution and count-rate capability of a germanium detector would be far superior, germanium detectors were not used in the subject applications due to cost, fragility, and bulky cooling apparatus. Due to the high atomic number of CdTe and CdZnTe detectors, the peak-to-total ratio is greater than with small germanium detectors, while the peak-to-Compton ratio is similar.

II. RESULTS AND DISCUSSION

A. HLWT Environment

Since 1943, 176 single- and double-shelled underground storage tanks were constructed and filled with highly radioactive chemical waste. The tanks are constructed with carbon steel lining the bottom and sides of a reinforced concrete outer shell (Fig. 1). Waste tanks typically are 23 m (75 ft) in diameter and have capacities of up to 3,790,000 liters (1,000,000 gallons). The materials contained in the HLWTs are byproducts from nuclear fuel reprocessing at the Hanford Site, primarily sodium hydroxide, sodium salts of nitrate, nitrite, carbonate, aluminate, and phosphate, and hydroxides of iron and manganese. Once transferred to the tanks, the wastes are maintained at highly alkaline conditions (pH 10-14).

The radioactive components consist primarily of fission product radionuclides such as ^{90}Sr and ^{137}Cs, activation products such as ^{60}Co, ^{152}Eu, and ^{154}Eu, and isotopes of actinide elements, such as uranium, plutonium, and americium. The ^{90}Sr and ^{137}Cs isotopes are the primary heat and radiation sources within the tanks. Fission and activation products generate gamma-ray dose rates up to approximately 11 Gy/h (1100 R/hr) in the tank interior. Short-lived radionuclides are not important contributors to the radioactivity, as no new fission products have been added since 1986 and most of the waste was generated in the decades prior. The predominant and large abundance of ^{137}Cs makes measurements of other isotopes with photon energies below 662 keV impossible in most circumstances. Measurements of higher energy gamma rays, ^{60}Co, ^{152}Eu, and ^{154}Eu, often provide significant information. These spectral features of the CdTe for an example waste tank are apparent in the spectrum shown in Fig. 4.

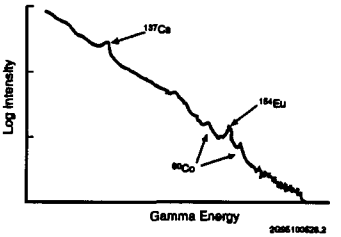

Fig. 4 *In situ* gamma energy spectrum from the bottom layer of an example HLWT

Measurement access is through penetrating drywells on the tank interior. Drywells are typically 7.6 cm (3 in.) inside diameter and were either installed during tank construction or were inserted after the tank was put into use. Isotopic profiles can only be acquired where drywells are available.

B. Tanks Containing Ferrocyanide Compounds

In early Hanford Site processing operations, liquid wastes were treated with ferrocyanide compounds to scavenge and remove or precipitate Cs isotopes. The highly active ferrocyanide compounds were transferred into tanks for long-term storage. The receiving tanks contained other wastes comprised of nitrate and nitrite compounds. Concerns have been raised about the potential for explosive ferrocyanide/nitrate reactions that could spread large amounts of activity over a wide area.

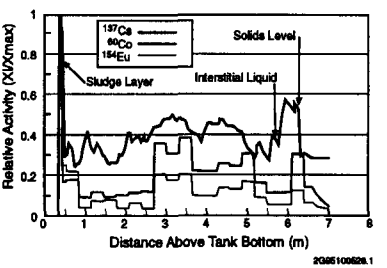

Fig. 5 Radioactivity profiles for HLWT 241 BY-106

As a prelude to sampling, remote characterization was performed in twelve of the ferrocyanide-containing tanks to determine the distribution of radionuclides within the tank. Profiles of activity within the tanks were generated. Fig. 5 depicts ^{137}Cs, ^{154}Eu, and ^{60}Co activity within the Hanford Site

tank 241-BY-106. In order to show inter-relationships, each isotope profile in the figure has been scaled to a maximum value of one. Large variations in activity that can occur over a very short distance can be seen. This information yields clues as to possible ferrocyanide complexed Cs distributions within the tanks and assists in planning for core sampling the tank.

C. Component Distribution Inference

Analytical results of two core samples from tank 241-AZ-101 established that ^{154}Eu is highly correlated to plutonium, ^{244}Cm, ^{60}Co, and ^{90}SrY activity within the tank. A high degree of correlation is also observed between ^{154}Eu activity and solids concentration in tank material. ^{154}Eu has a reasonably uniform concentration in tank solids and is essentially absent from the supernate. ^{137}Cs was found exclusively in the supernate and was not correlated at all to ^{154}Eu, plutonium, solids, ^{60}Co or ^{244}Cm.

Several measurements were taken within the tank. Collimated gamma-ray spectroscopy measurements show the distribution of ^{137}Cs, ^{60}Co, and ^{154}Eu throughout the tank. Passive total neutron counting with a 23-cm (9-in.)-long uncollimated BF_3 detector yielded a signal that was proportional to the neutrons in the tank which originate from contained transuranic elements, primarily arising from ^{244}Cm.

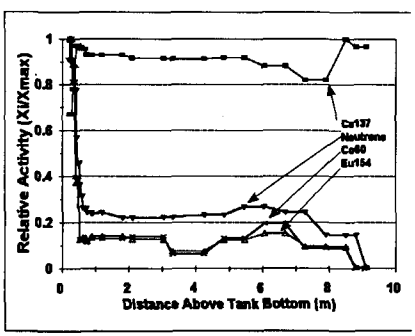

Fig. 6 Radioactivity profiles for HLWT 241 AZ-101

Fig. 6 depicts the relationship between ^{137}Cs, ^{60}Co, ^{154}Eu, and neutron activity within this tank. This profile is scaled as for Fig. 5 and corroborates the relationships predicted by core sample analysis between the different analytes. The distribution of neutrons is not as sharply defined as the gamma-ray measurements because the detector is long and uncollimated. However, the correlations show that initial assessments and continuing evaluations can be made in the waste tanks with the non-destructive assay used here.

D. Remote Hot Cell Application

Core sample analysis of tank waste is performed in Hanford Site hot cells. Core segments are extruded from the sampling device, and sub-sampled for laboratory analysis for a full suite of organic, inorganic and radiological analytes. After extrusion, a preliminary assessment of the core segment is performed using a remote measurement probe constructed for gamma-ray spectroscopy using a CdZnTe gamma-ray detector, and for beta spectroscopy using a silicon surface barrier detector (Fig. 7) [8].

This approach provides immediate assessment of the isotopic distribution within the core sample so that sub-sampling of the core sample can be focused on zones of interest. It is expected that the total number of samples analyzed by the laboratory will be reduced from more efficiently sub-dividing the core into sub-samples. Results are also used for health physics purposes in determining handling criteria of the sample once the sample is removed from the cell.

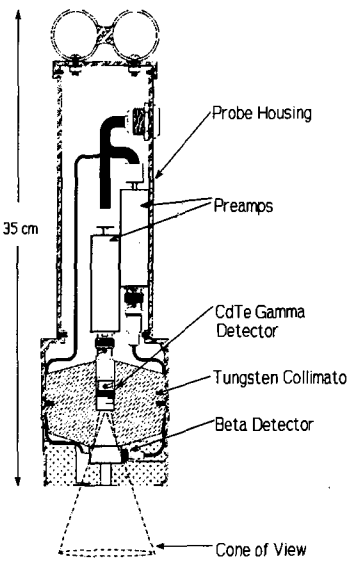

Fig. 7 Hot cell probe design with both gamma and beta spectroscopy detectors

III. FUTURE WORK

A. Tank Farms

It is planned that large mixer pumps will be installed into tank 241-AZ-101 in order to mix the waste into a slurry. In 1996 the pumps will be installed and operated to test this performance. It is planned that *in situ* gamma-ray spectroscopy will be performed through 8 in-tank drywells

during mixing to monitor the dispersion of solids. Work is underway to develop an assay system to support this effort.

B. Fuel Storage Basin Sludges

It is proposed to access the radionuclide content in accumulated sludges dispersed in the bottom of the 105-KE spent fuel storage basin. It is expected that a few physical samples will be taken of the sludge, and *in situ* gamma-ray spectroscopy will be performed at numerous locations at the bottom of the basin to yield a more detailed description of the sludge throughout the basin.

C. Instrumentation

It is desired to design new pulse processing electronics specifically for CdTe detectors. An empirically derived algorithm to more effectively compensate for charge trapping in planer CdTe and CdZnTe detectors has been developed and is described.[4]

IV. CONCLUSIONS:

The small size, room temperature operation, and the high atomic number of CdTe and CdZnTe gamma-ray detectors provide for a low-cost spectroscopy system that has proven useful in space-limited applications with high gamma-ray fields. Pulse risetime compensation has been developed to partially overcome the undesirable effects of charge trapping. A third-order polynomial fit of the background to the logarithm of the channel number provides a good means of determining the spectral background to photopeaks.

CdTe and CdZnTe gamma ray spectroscopy has been successfully used in: *in situ* characterization of high-level radioactive level waste storage tanks, analytical screening of samples within a Hanford Site hot cell, and general non-destructive assay projects.

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