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Hot Cell Remote Nuclear Scanning of Tank Core Samples

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Prepared for the U.S. Department of Energy
Assistant Secretary for Environmental Management

Westinghouse
Hanford Company Richland, Washington

Management and Operations Contractor for the
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Hot Cell Remote Nuclear Scanning of Tank Core Samples

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ABSTRACT

A Westinghouse Hanford Company (WHC)-designed remote measurement system has been constructed for gamma and beta isotopic characterization of Hanford Site high-level waste tank core sample materials in a hot cell. A small, collimated, planar CdZnTe detector is used for gamma-ray spectroscopy. Spectral resolution of 2% full-width-at-maximum at 662 kiloelectronvolts (keV) has been obtained remotely using risetime compensation and limited pulse shape discrimination (PSD). Isotopic measurement of high-energy beta emitters was accomplished with a ruggedly made, deeply depleted, surface barrier silicon detector. The primary function of the remote nuclear screening system is to provide a fast, qualitative stratigraphic assessment (with isotopic information) of high-level radioactive material.

The probe represents the first implementation of core scanning at the Hanford Site. Other approaches to real-time measurement are in the planning stage. Measurements by this method can indicate the degree of non-homogeneity within the core without costly subsampling of each portion of the core segment. A 5-point scan of a 48-cm core segment takes only 6 minutes.

Both gamma spectroscopy and beta measurements have been performed on actual core segments. Differences in radionuclide content, which correspond with color or texture variations, have been seen in constant cross section core samples, although for many samples the activity variation can be ascribed to geometry and/or mass factors. Discussion of the design, implementation, results and potential benefits will be presented.

INTRODUCTION

Laboratory analysis of highly radioactive, chemically complex materials is time consuming, hazardous, and extremely expensive. Analysis of high-level radioactive samples begins with sample preparation in shielded confinement chambers called hot cells. Analytical screening of samples within the hot cells can significantly reduce problems associated with mixed waste characterization. A remote screening probe for *in situ* measurements provides the following:

- Immediate assessment of isotopic distribution within the sample
- Focused analytical work on sample zones of interest
- Increased laboratory capacity by minimizing the number of analyses required per sample and minimizing reruns
- Warnings of samples that could present a radiological hazard or a contamination control problem.

A multidetector remote probe has been developed and deployed in a hot cell for screening purposes. This probe is an evolution of technology developed for *in situ* characterization of the high-level waste (HLW) tanks used to store high-level nuclear waste at the Hanford Site [1,2]. Probe mobility is provided by hot cell manipulator arms that grip handle rings. The probe is connected to the nuclear instrumentation on the exterior of the hot cell by two seven-core cables. While able to perform other tasks, the major function of the hot cell screening probe is to determine semi-quantitative isotopic distributions within HLW tank samples. This is accomplished without significant modification to the hot cells or sample handling procedure.

The isotopes selected for measurement were ^{137}Cs, ^{154}Eu, ^{152}Eu, ^{60}Co and $^{90}Sr/^{90}Y$. These isotopes were chosen because they constitute the majority of the activity in HLW tanks and are good indicators of material composition [1, 2, 3]. For example, ^{154}Eu is typically found with the insoluble transuranics while ^{137}Cs is usually found in aqueous regions or soluble salt layers.

Measuring uranium and transuranic radionuclides would also be valuable. Unfortunately, these are weak- or low-energy gamma-ray emitters; few are beta emitters. These properties preclude nondestructive assay of the actinide isotopes by beta or gamma-ray spectroscopy in the wet, dense, $^{90}Sr/^{90}Y$ and ^{137}Cs rich HLW tank material.

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Without sample contact, alpha spectroscopic measurements of HLW tank material are not viable. The expected neutron emission rate is very low and size limitations on the probe prohibit passive neutron measurements with reasonable efficiency. Notwithstanding the neutron count rate, the vast majority of neutron emissions in HLW tank material originate from the spontaneous fission of ^{244}Cm and alpha-n reactions from ^{241}Am, non-fissile isotopes with limited importance. For the above reasons, a measurement of actinide elements with alpha or neutron detectors was not incorporated into the probe.

Moisture content is a key safety parameter for the HLW tanks, and it is a high priority for analysis. Hot cell ventilation creates a substantial air flow past the samples, resulting in significant evaporation. To minimize the loss of water from the sample, counting times must be limited.

ENVIRONMENT

Nuclear Waste Material

The materials contained in the HLW tanks originate from treated process wastes, primarily sodium salts of nitrate, nitrite, hydroxide, carbonate, aluminate, and phosphate. Hydroxides of iron, manganese, and other transition metals are also present. Additional process wastes and remediation additives include di-2-ethylhexylphosphoric acid (HDEHP), ethylenediaminetetraacetic acid (EDTA), hydroxy-EDTA (HEDTA), tributyl phosphate (TBP), methyl isobutyl ketone (hexone), normal paraffin hydrocarbons (similar to kerosene with C_{10} to C_{15} hydrocarbon chains), diatomaceous earth, ferrocyanide complexes, and decomposition products of the foregoing.

These materials exist in the following general forms: sludges, saltcakes, and liquids. Sludges, located in tank bottoms, consist of solids (hydrous metal oxides) precipitated from the neutralization of acid wastes before their transfer to the HLW tanks. Once transferred to the tanks, the wastes are maintained at highly alkaline conditions (pH 10-14). Saltcakes are made up of salts formed from the evaporation of water from these alkaline wastes. Saltcake crusts may reside on top of the liquids. Liquids may exist as supernate and/or interstitial liquid.

Radioactive components consist primarily of fission product radionuclides, such as ^{90}Sr and ^{137}Cs; and actinide elements, such as uranium, plutonium, and americium. The strontium and cesium isotopes are the primary heat and radiation sources within the HLW tanks, generating up to seven gray per hour (700 Rads per hour) dose rate on contact. At this time, short-lived radionuclides are not important contributors to the radioactivity because no new fission products have been added since 1986, and most waste was generated in the 1950s and early 1960s.

A number of sampling types support HLW tank characterization [4]. The HLW tank solid sampling methods include supernate grab sampling, auguring, and rotary- and push-mode coring. Supernatant sampling is used for liquids, light solids, and slurries. Auger samples are taken by an auger screw; they consist of materials with the top 40 cm from the top of the waste sampled. Auger sampling is unsuited to very dry or slurry-type samples; it is used only for saltcake-type crust materials. An auger sample loses stratigraphic information because there may be some mixing during the sampling process. The only methods that provide a continuous sample throughout a vertical cross section of the waste tank are rotary- and push-mode sampling. Both provide sequential 2.5-centimeter (cm) diameter by 48-cm long samples (called segments). A push-mode sampler is a "thief" style coring device; it is suited to softer sludges. The rotary-mode sampler drills through hard saltcake materials that are present in some tanks.

A core segment comes to the laboratory encased in several layers of shielding and containment. The hot cells are the place where the core is removed from containment, and the segment is subsampled for analysis. A core segment is extruded by being pushed out of the sampler onto a moving tray. The core segment lies on the tray in the stratigraphic order that it had in the sampler and presumably had in the tank. Depending on the cohesiveness of the core segment, it can retain its cylindrical shape, or it can slump flat. The colors range from off-white to brown (most common) to black. The segment is subsampled by determining differences in segment strata or differences from adjacent core segments.

The HLW tank contains every element in the periodic table (as fission, neutron activation products or process materials) and a wide range of molecular and ionic species (inorganic and organic) from nuclear fuel processing. Laboratory analyses of HLW tank material include a variety of analytical techniques [4]. A short list of principal analyses includes the following: inductively coupled plasma atomic emission spectroscopy for most metals and a few nonmetals; ion chromatography for anions, differential scanning calorimetry for heat-producing or absorbing qualities of the waste; volatile organics, semi-volatile organics, gamma energy analysis for gamma ray emitting radionuclides; and total beta and alpha energy analysis. A wide variety of other chemical, physical, and radiological analytical techniques are used to determine the properties and hazards of a specific sample.

A single HLW tank usually has one or two complete core samples taken from it because of the high cost of sampling and the associated analyses. Hot cell sample screening reduces or eliminates many expensive, hazardous, time-consuming, and redundant laboratory analyses.

Hot Cells

Difficulties in applying technology to remote hot cell characterization are related to remote operation in harsh radioactive and caustic chemical environments. Instrument probes in a hot cell environment must be impervious to prolonged radiological exposure, corrosive vapors, and splashing from normal hot cell cleaning procedures. Radiation background is normally less than 0.1 milliroentgen per hour (mR/hr) outside the hot cell. Within the hot cell, radiation levels vary from 500 mR/hr to 10 R/hr, with the potential to exceed 100 R/hr when higher radiation level tank cores or other samples are being processed. Chemical contaminants of primary concern are condensed vapors, liquids, and solids (pH from 8 to 14), and cleaning agents that contain six Molar nitric acid.

Current Sample Preparation Methods

Core segments are prepared for analysis by compositing and homogenizing whole- or half- segment subsamples (or more rarely, quarter-segment subsamples). The subsamples are removed from the hot cell and put through a digestion/ dilution preparation prior to actual analysis. Gamma energy analysis and total beta samples are mounted on a flat dish for counting. ^{90}Sr samples are put through a separation process, then are mounted and counted.

EXPERIMENTAL

Multi-Instrument Remote Probe

The harsh, confined environment within hot cells requires a probe that is as rugged and compact as possible. Because the cabling required a controlled penetration of radiation zones, the number of cables and connection sizes was also reduced to a minimum. Several probes of different configurations have been constructed. The weight and size of the probe had to be minimized to allow remote manipulator arms to provide mobility and positioning. Figure 1 shows the hot cell probe, which weighs only 10 kilograms. The probe position is determined by using a pair of laser-line generators mounted in the probe bottom. Mounted and aligned orthogonally to each other, these 60-milliwatt diode laser-line generators produce a red X on a sample. The center of the X is axially aligned with the probe for unambiguous positioning of the probe over any surface.

Preamplifiers for both detectors are mounted within the probe head, and all other signal processing equipment is externally located to the hot cell. The preamplifiers used for both and beta systems are EuroRad[b] PR6 models, chosen for their performance, small size, and compact connections.

The type of cabling and connectors that will work in a hot cell environment are constrained by small openings in the hot cell, resistance to chemical attack, and radiological contamination concerns. Separate multiconductor marine

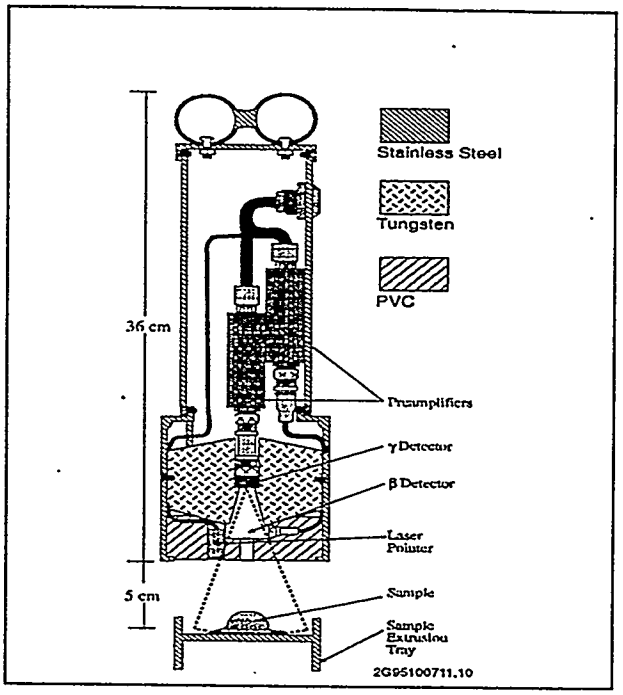

Fig. 1 Remote Hot Cell Probe.

video cables were used for the beta and gamma detectors. Each cable consists of two coaxial cables, five single conductors, and an overall metal foil shield. Environmentally sealing LEMO[c] quick couplers are used at both ends of the cabling that penetrate the hot cell wall. This allows the removal of the probe head from the hot cell and enables the instrumentation rack outside the hot cell to be disconnected and moved away from other projects. Careful attention to grounding was required to prevent ground loops, crosstalk, and ringing.

To prevent cross-contamination of the samples and allow positioning, the probe stand-off distance from the sample is approximately 5 cm. All collimators were designed and experimentally shown to be radially symmetric. Probe design provided insignificant beta interference on the gamma detector. Gamma interactions with the silicon beta detector were unavoidable. It should be noted that the silicon detector used is capable of alpha spectroscopy as well, but this capacity was not exercised for reasons discussed previously. The beta detector was made rugged and gold plated to function reliably once deployed. A 0.2-millimeter (mm)-thick aluminized mylar

[b]EuroRad is a trademark of the EuroRad Corporation, Strasbourg, France.
[c]LEMO is a trademark of LEMO USA Inc., Santa Rosa, California.

window was placed below the beta detector to prevent ambient light interference and for physical protection from corrosive splatter. Polyvinyl chloride was used as a lightweight, durable beta collimator on the probe bottom.

The requirements on the gamma-ray collimator are the principal defining constraints for the probe's size, shape, and weight. Reduced collimation or shielding requirements result in smaller, lighter probes. A machinable high-density tungsten alloy was chosen as the collimation material because it provides superior gamma attenuation per unit volume. Using Monte Carlo Neutron Photon (MCNP) modeling, two collimators were designed for measuring isotopic distribution within samples. The high-resolution collimator had a spatial resolution of approximately ± 2.5 cm; the low resolution collimator had a spatial resolution of approximately ± 4 cm. The high-resolution collimator requires significantly longer counting times, and more measurements must be taken to profile a sample. The lower-resolution collimator provides shorter data acquisition times but is less sensitive to spatial variations in the sample. The low-resolution gamma collimator was selected for installation in the hot cells.

The spatial collimation for beta detection is much tighter, about ± 0.5 cm at a typical sample standoff distance. However, the low sheilding capability of the collimation material (polyvinyl chloride) for gamma rays means that the beta detector responds to gamma rays over a wide-angle. This wide-angle response decreases the effective resolution for the beta detector, except for purely beta-emitting sources or samples.

Gamma Spectroscopy

Gamma-ray energy spectroscopy provides some of the most useful information about mixed waste. Spectral data allow gamma-emitting radionuclide content and distribution to be determined. However, the application for *in situ* spectroscopic measurements of high-radiation fields with a remote compact probe places unusual demands on the instrumentation.

Traditional high-resolution germanium gamma-ray detectors require cumbersome cooling and are expensive. Room temperature scintillation detectors yield low-resolution spectra that limit isotopic identification. CdTe and CdZnTe detectors, with electronic signal processing enhancements, were selected for this application. The specific advantages of the high-Z semiconductor detectors include the following:

- Small size
- Room temperature operation
- High-Z material (provides good stopping power and moderate resolution)
- Low cost
- High count-rate operation.

Charge trapping within high-Z semiconductor detectors significantly degrades spectral resolution. Pulse risetime compensation (PRC) is a technique used to compensate for charge trapping by adjusting the amplitude of the detector pulse by an amount depending on the risetime [2, 5, 7, 8, 9]. A small percent of pulses, with extremely long risetimes originating from events with a high degree of charge trapping, are rejected using pulse shape discrimination (PSD) [6].

The signal processing electronics for the gamma system were composed of five commercially available nuclear instrument modules between the preamplifier and the multichannel buffer. The system has two major sections: compensation and discrimination. The compensation section consists of a shaping spectroscopy amplifier used in conjunction with an EG&G ORTEC[d] Model 675 Ge resolution enhancer. The resolution enhancer adjusts the amplitude as a function of preamplifier pulse risetime. Pulse risetime in CdTe and CdZnTe is a function of the degree of charge trapping that occurs within the detector. The corrected pulses are sent to a multichannel buffer for spectral analysis.

The discrimination electronics are made up of a delay line amplifier, a pulse shape analyzer/timing-single channel analyzer, and a time-to-amplitude converter/single channel analyzer. These modules sort out pulses with long risetimes that originate from pulses with a large degree of charge trapping within the detector, and which cannot be effectively compensated by PRC circuitry. PSD electronics produce a coincident gate pulse that enables counting only the "good" pulses in the multichannel buffer.

Using PRC, PSD, and high bias on the detectors, excellent performance was extracted from the small high-Z semiconductors. CdZnTe spectral resolutions of better than 2.0% full-width-at-maximum at 662 keV have been obtained with a 200-V bias, PRC, and rejection of approximately 10% of the longest rising pulses with PSD. This resolution is a significant improvement over unenhanced crystals and is better than the standard sodium iodide scintillators. It is not as good as conventional germanium semiconductor detectors. With the improved spectral resolution, deconvolution of the gamma photopeaks is not necessary for analysis.

The gamma measurement system has a secondary mode of operation for the rapid acquisition of nonspectroscopic

[d]EG&G ORTEC Inc. is a trademark of ORTEC Incorporated, Oak Ridge, Tennessee.

data. Single-channel analyzers and counter/timers were used to collect data from selected windows efficiently. The windows selected were total gamma and total gammas above ^{137}Cs. Total gamma provides a measurement of the total gamma radiation from the sample, which is primarily ^{137}Cs in HLW tank material. Total gammas above ^{137}Cs accumulates all signals above the ^{137}Cs photopeak and provides a high-efficiency, nonselective measure of the isotopes ^{154}Eu, ^{152}Eu, and ^{60}Co.

The key parameters for the hot cell detector were efficiency (allowing shorter counting times) and spectroscopic resolution for isotopic identification. The detector selection for the hot cell probe was made by comparing data from nine different CdTe and CdZnTe detectors with the same source, geometry, and instrumentation settings. Manufacturing variance is large enough that detectors are individually selected for each application, but general trends were observed. Smaller crystals were found to have better spectroscopic resolution. It is postulated that this is caused by reduced charge trapping. Some crystals tested were cylindrical; others were cubic. The crystal geometry did not seem to impact resolution. As expected, larger crystals, and those of CdZnTe, had higher efficiency. A 5-mm x 5-mm x 1-mm CdZnTe detector was selected as the best for this application.

Beta Spectroscopy

A small, ruggedly made, 25 square millimeter deeply depleted (5,000 microns) silicon surface barrier detector was chosen to perform *in situ* beta spectroscopy within the hot cell. The deep depletion zone was required to capture the high-energy fission product beta particles. The beta spectroscopy system that uses a typical spectroscopy amplifier and a fast multichannel buffer. Additionally, the signal is split into two single-channel analyzers, and the output was directed to the timer/counters. The single-channel analyzers efficiently collect two data channels from the detector: the silicon detector signal and the total betas above gamma.

The isotopic information obtainable from a beta spectrum is more limited than the photopeak information available in a gamma-ray spectra. Although more complex than gross counting, beta spectroscopy is preferred because it provides information for system diagnostics, and isotopic information can be more correctly inferred from spectrum shape. The air gap between sample and probe and the variances in sample moisture content and geometry affect the beta spectrum.

A measurement of the total beta signal above the energy of the gamma-ray responses for photopeak interactions from the 1,332-keV gamma ray of ^{60}Co provides a measurement of ^{90}Y, the daughter of ^{90}Sr. This measurement is called total betas above gamma. In aged fission product waste, the only significant interference is from ^{234m}Pa, a daughter of ^{238}U. However, uranium recovery efforts from the fission waste have left little uranium to generate the ^{234m}Pa daughter. Insignificantly small interferences originate from pulse pile-up, ^{152}Eu and ^{154}Eu, and the natural radionuclides, including ^{40}K and the decay products of uranium and thorium. The isotopes ^{106}Rh (daughter of 368-day ^{106}Ru), ^{144}Ce (284-day) and its daughter ^{144}Pr have beta particles with interfering energies. For all practical purposes, these isotopes have decayed away because the last Hanford Site reactor was shut down in 1986; and if they were present, they would be noted in the gamma-ray spectrum. Other isotopes have decayed away, are present only in trace quantities, or have beta and gamma-ray energies that do not cause concern. The beta spectral shape from $^{90}Sr/^{90}Y$ is visually distinctive and readily distinguishable from all interferences except ^{234m}Pa. In aged high-level radioactive waste, the signal in the silicon surface barrier detector with energies above the response from ^{60}Co is strong and overwhelming from $^{90}Sr/^{90}Y$ activity.

The silicon detector signal measurement is actually a combination of high-efficiency beta and low-efficiency gamma interactions within the silicon suface barrier detector. The signal is a very crude indication of total radiative beta and gamma emissions from the sample. Of all subject radiometric measurements, it is the most sensitive measurement to low levels of activity.

CALIBRATION, QUALITY ASSURANCE, DATA ANALYSIS

The hot cell measurements are semi-quantitative. Extruded samples typically have irregular geometries. The instrument was calibrated by measuring ^{60}Co, ^{137}Cs, and ^{90}Sr standards that approximate sample geometries. The standard sources had two configurations: square tray and short cylinder. The shapes were chosen to mimic the extremes of the geometry of the samples; that is, flat slush in a sample tray or round "logs." Because the sources were not long enough to mimic a complete core segment, selected sources were counted at several positions out from the center viewing area of the detector. The resulting counts in the region of interest were later added together to simulate a complete core.

The instrument is checked for drift in energy and efficiency by taking spectra of a ^{154}Eu source before and after each calibration count and each day before the analysis of samples. A $^{137}Cs/^{90}Sr^{90}Y$ source is also available but not normally used in daily operations.

A minimum detectable concentration is defined by the Hanford Analytical Services Quality Assurance Plan (DOE/RL-94-55) [10] and is shown in the following equation:

$$MDA = \frac{2.71 + 3.3\sqrt{(R_b * T_b) * \left[1 + \frac{T_b}{T_t}\right]}}{e * b * LT * k}$$

where

R_b = background count rate
T_b = background count time
T_t = sample count time
e = counting efficiency
b = abundance
L_T = elapsed live time (background counting time = T_b)
k = 37,000 disintegrations/second/μCi.

In this application, values k, b and e are combined with a concentration term, due to the nature of the standards. This concentration only applies to the measurements of ^{137}Cs and ^{90}Sr.

The minimum detectable activity level is given in counts per second for the total beta, total gamma, and gammas-above-cesium measurements.

RESULTS AND DISCUSSION

Using pulse risetime compensation and limited pulse shape discrimination, the 5-mm x 5-mm x 1-mm planar CdZnTe detector with the remote probe provided spectral resolution of 2% full-width-at-maximum at 662 keV, the ^{137}Cs gamma-ray. Figure 2 shows the gamma energy spectra acquired with the remote probe of the calibration sources.

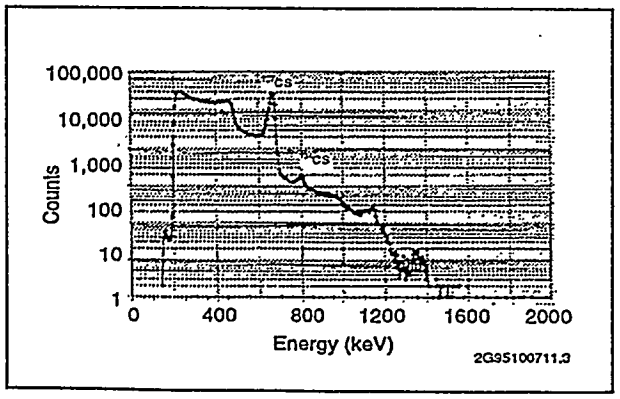

Fig. 2 HLW-Tank (241-AZ-102) Material Gamma Energy Spectra (6000 Seconds).

[e]GammaVision is a trademark of ORTEC, Inc., Oak Ridge, Tennessee.

The typically 60-second count time was sufficient to collect good spectra from the principle isotope ^{137}Cs. The small size of the detector and the tungsten shielding alleviated high count problems that might have been encountered, but longer count times were required to resolve trace isotopes such as ^{134}Cs, ^{154}Eu, ^{152}Eu, and ^{60}Co.

The one standard deviation, minimum detectable activity for the standard 60 second count time, varies as the background in the hot cell varies but is typically 1.6 μCi/g for ^{137}Cs, 2.7 counts per second for total gammas, and 0.3 counts per second for total gammas above ^{137}Cs.

The efficiency of the beta detector produced extremely high count rates on the HLW tank material. The computer (GammaVision)[e] multichannel buffer communication pathway could handle count rates up to approximately 90,000 counts per second. However, sample activity often exceeded 90,000 counts per second, and the initial calibrations had to be abandoned. Lower-level discrimination was adjusted upwards to reduce count rate.

Beta sensitivity was not an issue with the high efficiency of the detector and the high activity of the samples. However, the minimum detectable activity for 60-second count times was calculated to typically be 2.3 counts for the silicon detetctor signal and 0.15 μCi/g for total betas above gamma.

The extruded HLW tank core material rarely make ideal cylinders, but the calibrations on such cylinders are the most reasonable approximation.

Tank BY-110 Core 101

Tank BY-110 was chosen because it was being used as a test tank for a new rotary mode sampler, and several full core samples (seven to nine 48-cm core segments per full core) were planned for this tank. Segments 6a through 9 (of 9 segments) of core 101 were analyzed and five measurements per segment were taken. Figure 3 shows the relative values total beta, total gamma, ^{137}Cs, ^{90}Sr (betas above gammas), and gammas above cesium as a function of distance from the bottom of segment 9 (which also corresponds with the tank bottom).

The collected scans of tank BY-110 show that the sample becomes "hotter" as it gets closer to the tank bottom. This result is expected because the sample is more finely grained (thus having better packing and more mass) and because ^{90}Sr is more likely to be found in the solids.

A comparison of the tank BY-110 laboratory results to the probe scanning is not possible because the analyses have yet to be started (as of October 24, 1995) for the samples involved (extruded September 8, 1995). This

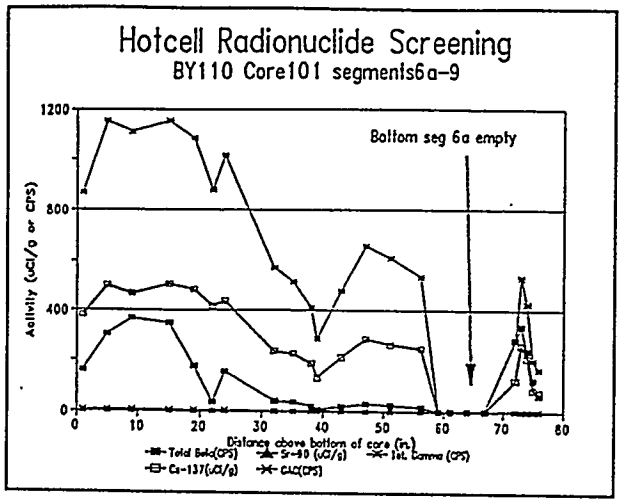

Fig. 3 Profile of BY-110 Core 101.

demonstrates the difference between the probe system and conventional methods in the time required for generating similar data.

Core Profile - Tank BX-103

Tank BX-103 has a much shallower depth of waste than tank BY-110; therefore, it had a full core with only two 48-cm segments. The results (Figure 4) clearly show the varying amount of mass underneath the probe for the top segment (segment 1, 10 cm and above), with hot spots corresponding to small clumps of material. The bottom segment was nearly uniform in cross section, so there were no great variations in mass to cause changes in activity. The higher activity in the case of segment 2 seems to be associated with dark material, as both ends of the segment were nearly black in color. Note that the material in segment 1 is all black in color.

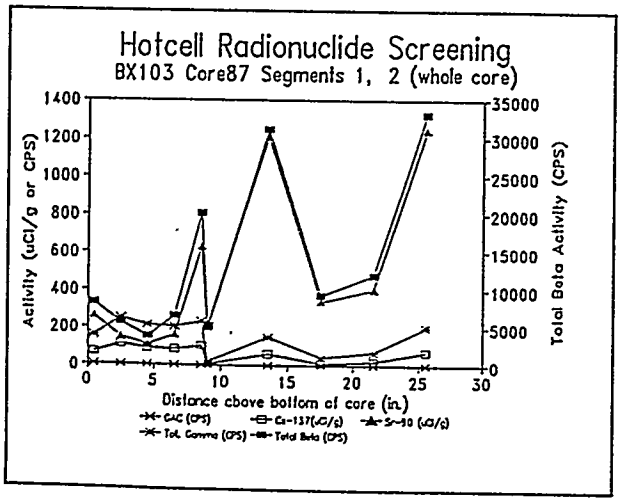

Fig. 4 Profile of Core 87, BX-103.

CONCLUSIONS

A remote semi-quantitative isotopic mapping system has been developed and demonstrated in a hot cell facility.

With customized pulse risetime compensation and pulse shape discrimination, signal processing gamma energy spectra have been taken with a remote CdZnTe detector that have 2.0% full-width-at-maximum resolution at 662 keV. Using the CdZnTe system, remote measurement of fission isotopes such as ^{60}Co, ^{134}Cs, ^{137}Cs, ^{154}Eu, and ^{152}Eu has been accomplished.

Small, ruggedly made, deeply depleted (5,000 microns) silicon surface barrier detectors were chosen to perform *in situ* beta spectroscopy within the hot cell. Isotopic and gross beta measurements were taken from fission wastes. $^{90}Sr/^{90}Y$ was the principal isotope.

Remote analytical screening of nuclear materials in hot cells provides information on the sample material in near-real time. Real-time data provides intuitive and objective information for preliminary sample assessment, direction of sub-sampling efforts, resolution of safety issues, and material transportation. This technology has other potential applications, such as process monitoring in nuclear power and fuel processing plants, safeguards, or remote characterization at mixed-waste remediation sites.

FUTURE WORK

For the discussed applications, chemical information would be as valuable as radiological information. Efforts are underway to produce a single integrated remote analytical system capable of collecting both isotopic and chemical information. Methods being investigated to augment the existing nuclear probe include forms of fiber optic spectroscopy [11]: Raman, Fourier transform infrared and fluorescence, time resolved laser-induced fluorescence, laser-induced breakdown (LIBS) and laser ablation mass spectroscopy (LA-MS). A small charge-coupled device camera could be incorporated into the probe to provide visual documentation of the measurement site. Finally, data fusion methods would condense the information from the probe's instrument array, allowing an organized, near-real-time look at multiphase materials in a hazardous environment.

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Hot Cell Remote Nuclear Scanning of Tank Core Samples

Waste Management '96

Shane Addleman Mark Beck Brian Keele Gordon Blewett
Gary Troyer

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Problem:

- Defense Nuclear Facilities Safety Board Recommendation 93-5
  - DOE/RL 94-0001 Commitment, 5.1, 5.4
- Core profiling
  - Hot spots, inhomogeneity, speciation
- Immediate data
- Lab needs
  - Subsampling guidance
  - Radiological warning

Hot Cell Environment:

- High radioactivity
- High contamination levels
- Corrosive atmosphere
  (over periods of months/years)
- No access for personnel
- Movement/adjustment limited by manipulator
- Time limitations
  (operations, sample)

Probe:

- Body is stainless steel
- Contains only detectors and preamps
- All other electronics in instrument rack
- Connectors are sealed quick disconnect
- Requires no physical adjustments
- Oriented for manipulator use

- Shielding:

  Gamma- Tungsten, shaped by modeling

  Beta - PVC

Detectors:

- Gamma detector
  - Solid state detector
  - Cadmium Zinc Telluride (CdZnTe)
  - Small
  - Room temperature
  - Medium resolution (better than NaI, not as good as HpGe), high atomic number (better photopeak/Compton for small volume)

Detectors (continued):

- Beta Detector
  - Solid state detector
  - Deeply depleted (optimized for $^{90}Sr/^{90}Y$) silicon barrier
  - Small
  - Room temperature
  - Ruggedized (vs shock, corrosion)
  - Transmission mount allows gamma ray pass-through

Scanning Approach:

- Simple operation
  - Use devices already in hot cell, no "single use" items
  - Complicated, breakdown-prone positioning equipment avoided
- Relative profile
- Indication of homogeneity
- Correlation of appearance vs component

Scanning Method:

- Blank taken before and after scan
- Sample moved underneath probe
- Distance from bottom of segment recorded
- Laser line cross hairs indicate location
- 1 minute count per location -
  5 positions per segment
- Data acquisition software automatically
  saves data

Data Reduction:

- Entire data reduction process < 10 minutes per core segment

- Target Components:

Gamma	Beta
^{137}Cs,	^{90}Sr
Total Gamma,	Complete Beta Detector
Gammas Above	Signal
Cesium Energy,	
^{60}Co, ^{154}Eu	

Data Reporting

- Performed at Hot Cell

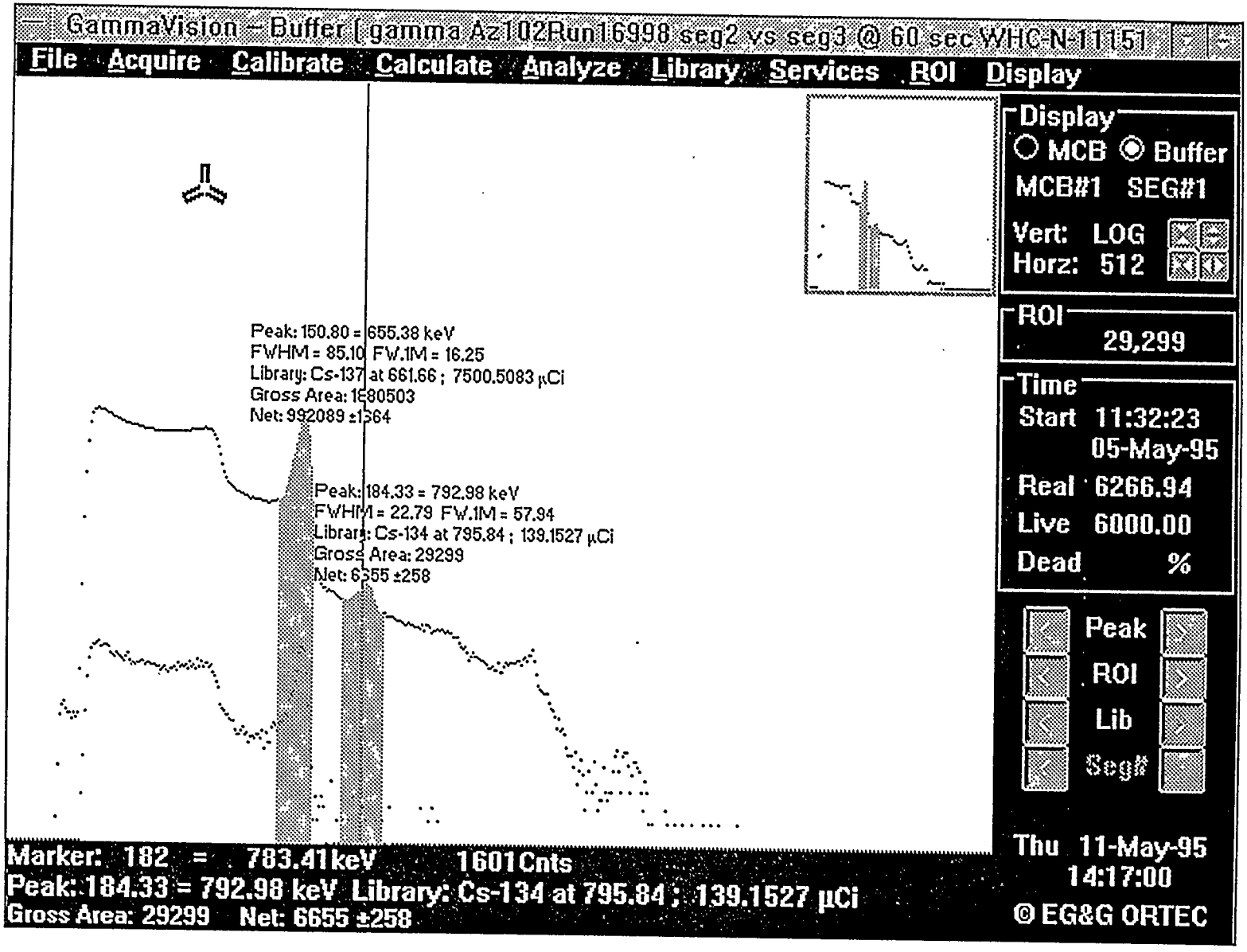
- Commercial software packages
  - Data Acquisition / Analysis - GammaVision
  - Data Reduction - Quattro Pro

- Reports
  - Tabular
  - Profile Charts

Initial Results:

BX-103: Actual push-mode core segment
Radiological variations within segment
Corresponds to visual features

Several archive samples (in vials) examined.
AZ-102: ^{134}Cs detected despite short half-life
AZ-101: much like AZ-102 (^{137}Cs and ß), simulated a non-homogenous segment.
U-201: very low activity seen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File Acquire Calibrate Calculate Analyze Library Services ROI Display
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MCB Buffer
MCB#1 SEG#1
Vert: LOG
Horz: 512
ROI
29,299
Time
Start 11:32:23
05-May-95
Real 6266.94
Live 6000.00
Dead %
Peak
ROI
Lib
Seg#
Peak: 150.80 = 655.38 keV
FWHM = 85.10 FW.1M = 16.25
Library: Cs-137 at 661.66 ; 7500.5083 µCi
Gross Area: 1680503
Net: 992089 ±1564
Peak: 184.33 = 792.98 keV
FWHM = 22.79 FW.1M = 57.94
Library: Cs-134 at 795.84 ; 139.1527 µCi
Gross Area: 29299
Net: 6655 ±258
Marker: 182 = 783.41keV 1601Cnts
Peak: 184.33 = 792.98 keV Library: Cs-134 at 795.84 ; 139.1527 µCi
Gross Area: 29299 Net: 6655 ±258
Thu 11-May-95
14:17:00
© EG&G ORTEC

Limitations:

- No alpha emitters (transuranics)
  Sample self absorption, standoff distance

- Non-quantitative

- Exposed core scanning time limited
  (to less than hours) by humidity/ drying issues

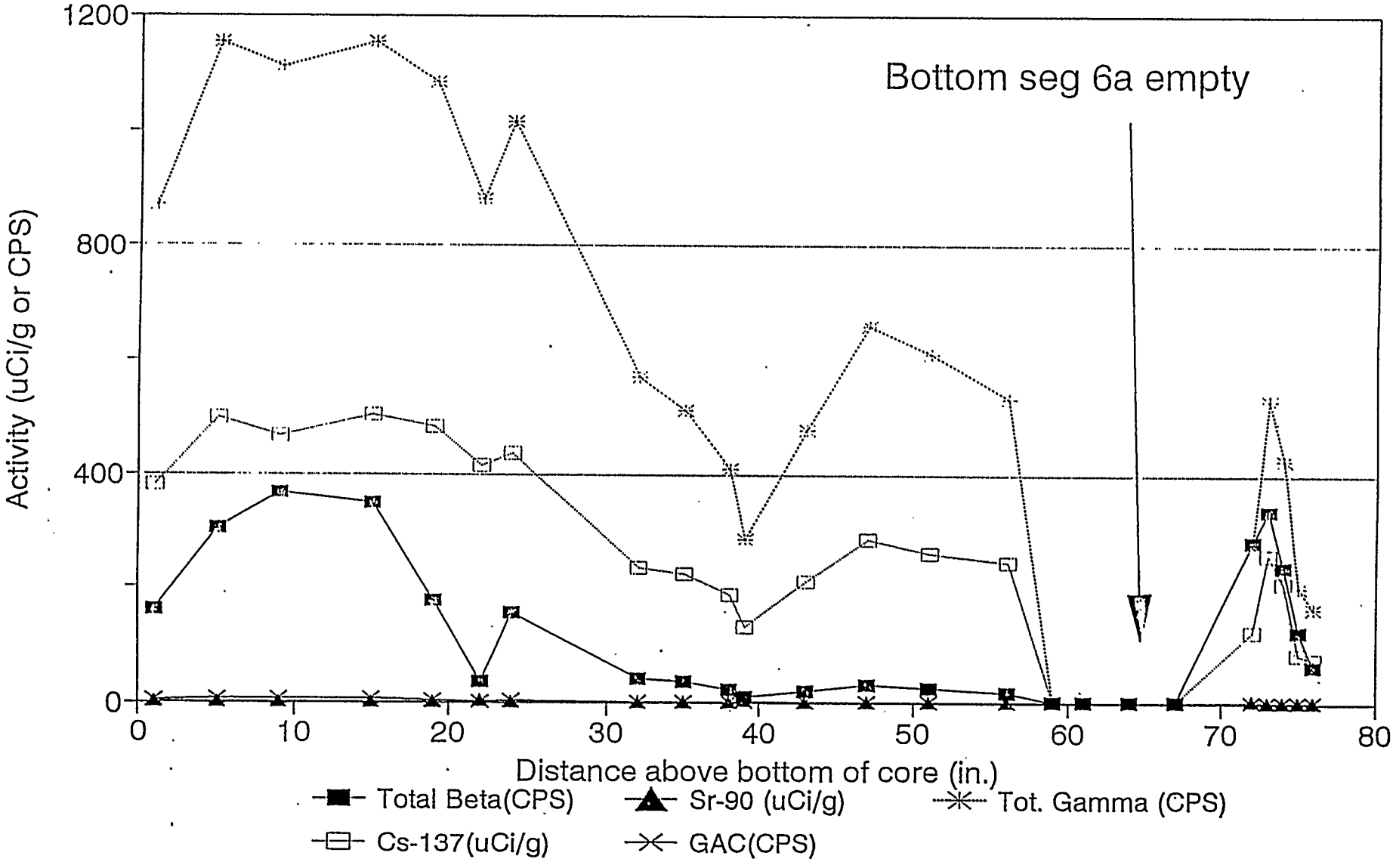
Benefits and Conclusions:

Hot Cell Screening Probe Provides:

- Stratigraphic information
- Immediate data, device installed, ready
- Subsampling guidance
- Radiological hazard warning

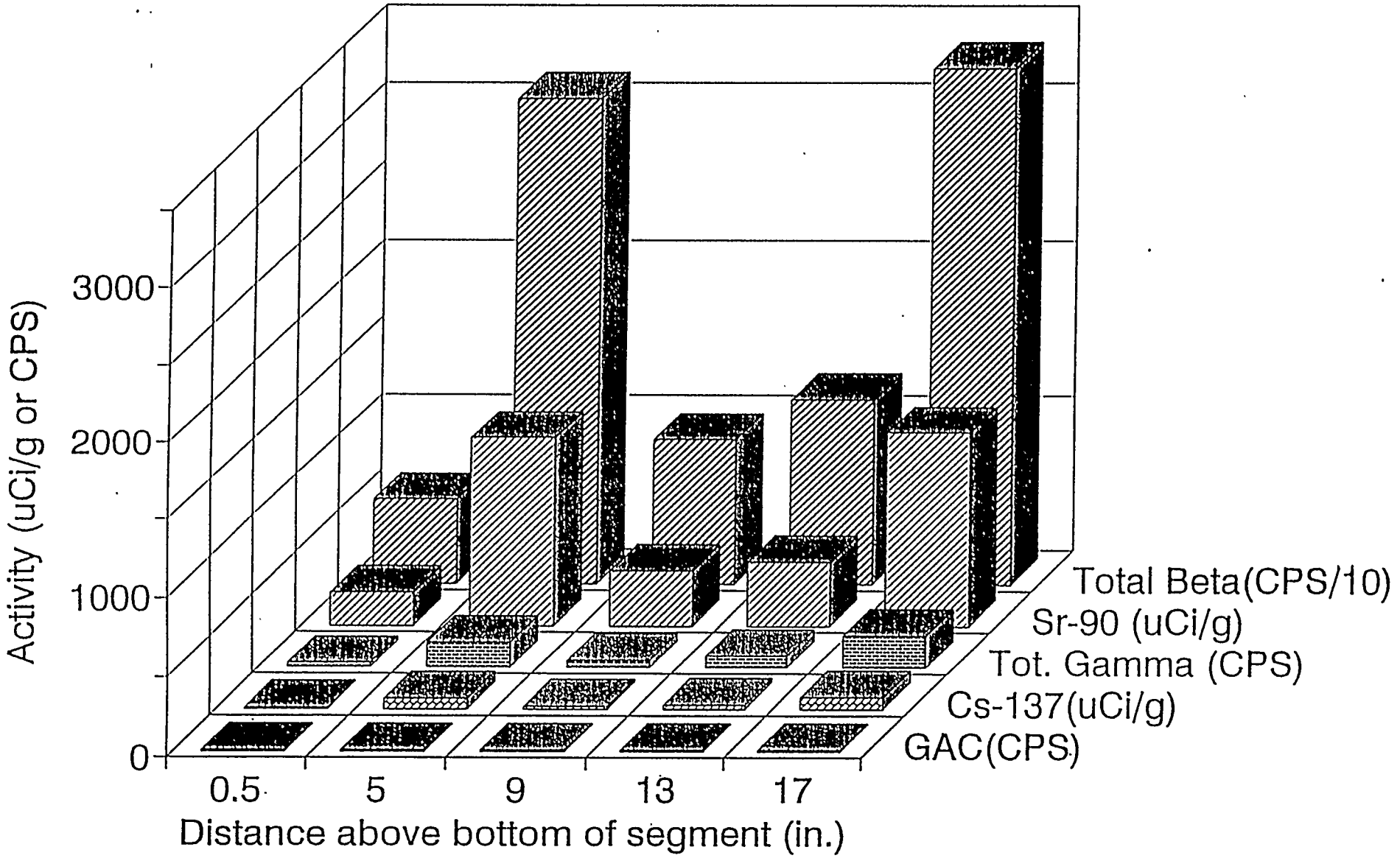
Hotcell Radionuclide Screening

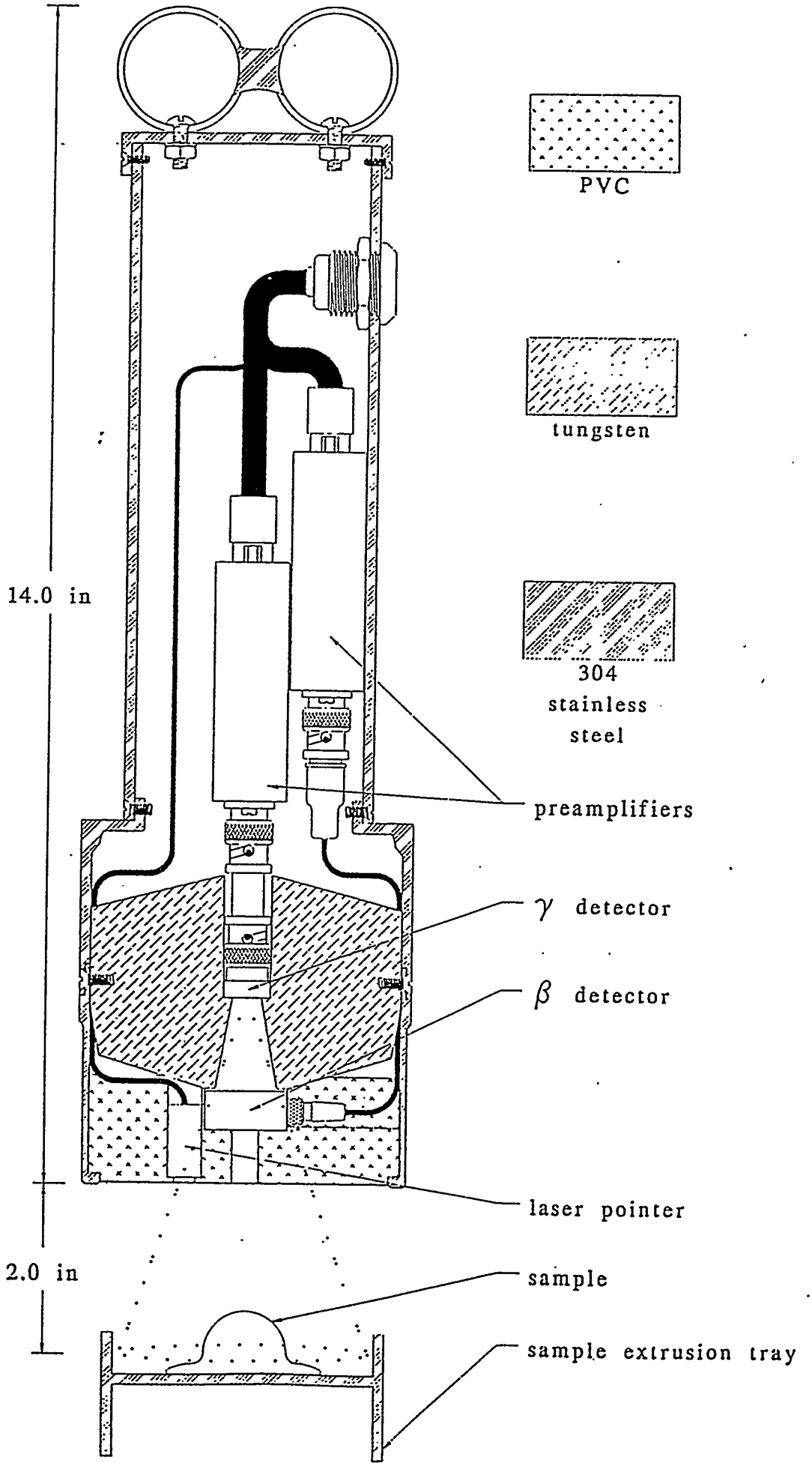
BY110 Core101 segments6a-9



Hotcell Radionuclide Screening

Core Segment:BX103 Riser#2 segment1



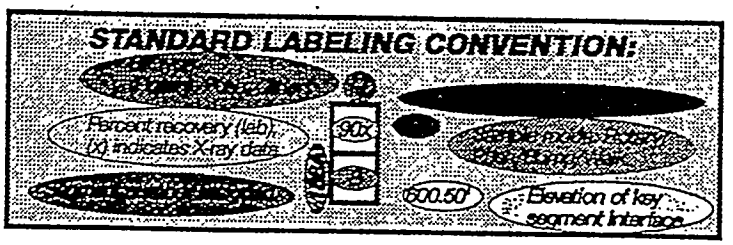

PVC
tungsten
304
stainless
steel
14.0 in
preamplifiers
γ detector
β detector
laser pointer
2.0 in
sample
sample extrusion tray

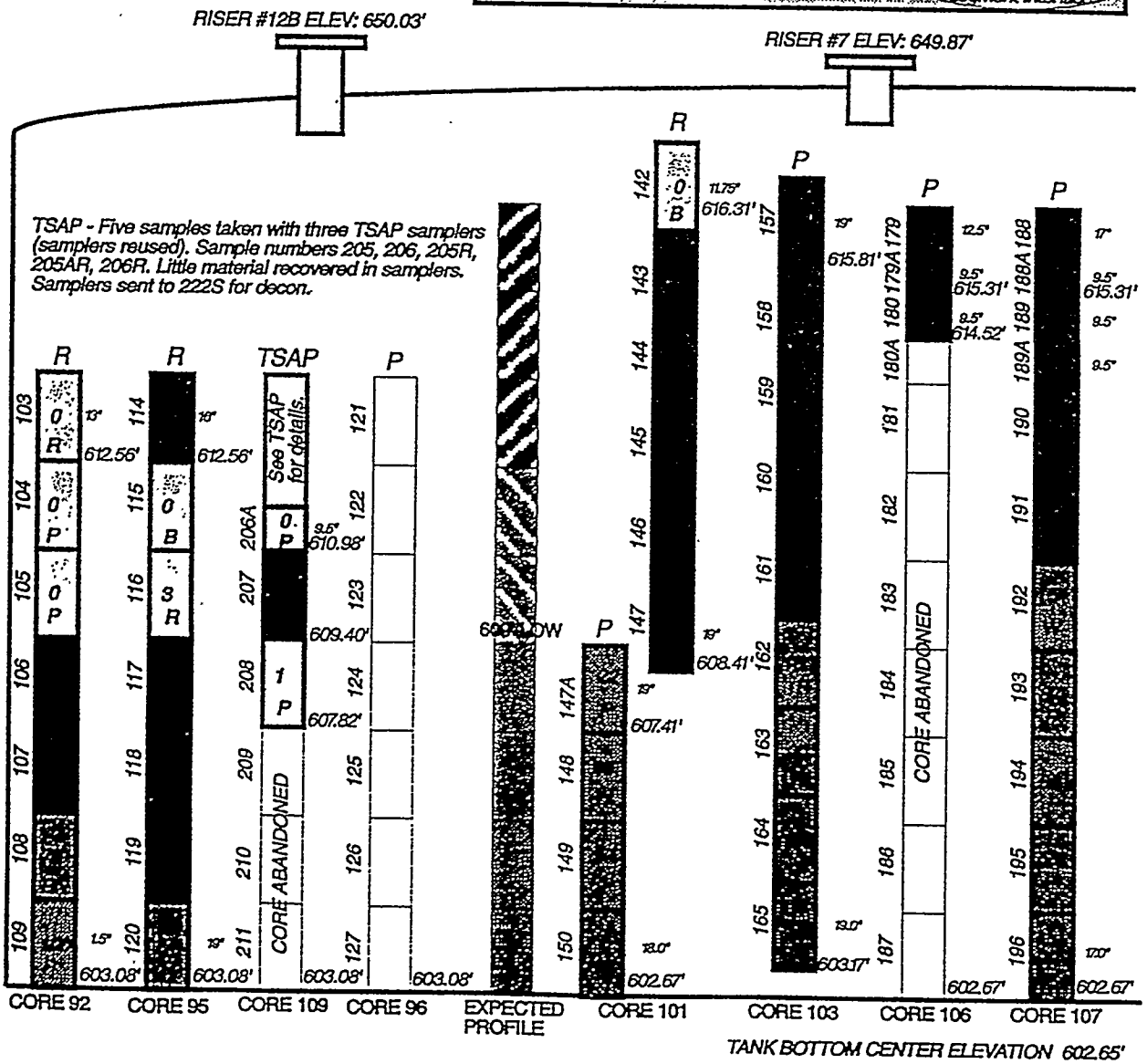
BY-110 RMCS CORE PROFILE

FILE: BY110.CRD as of 09/24/95.

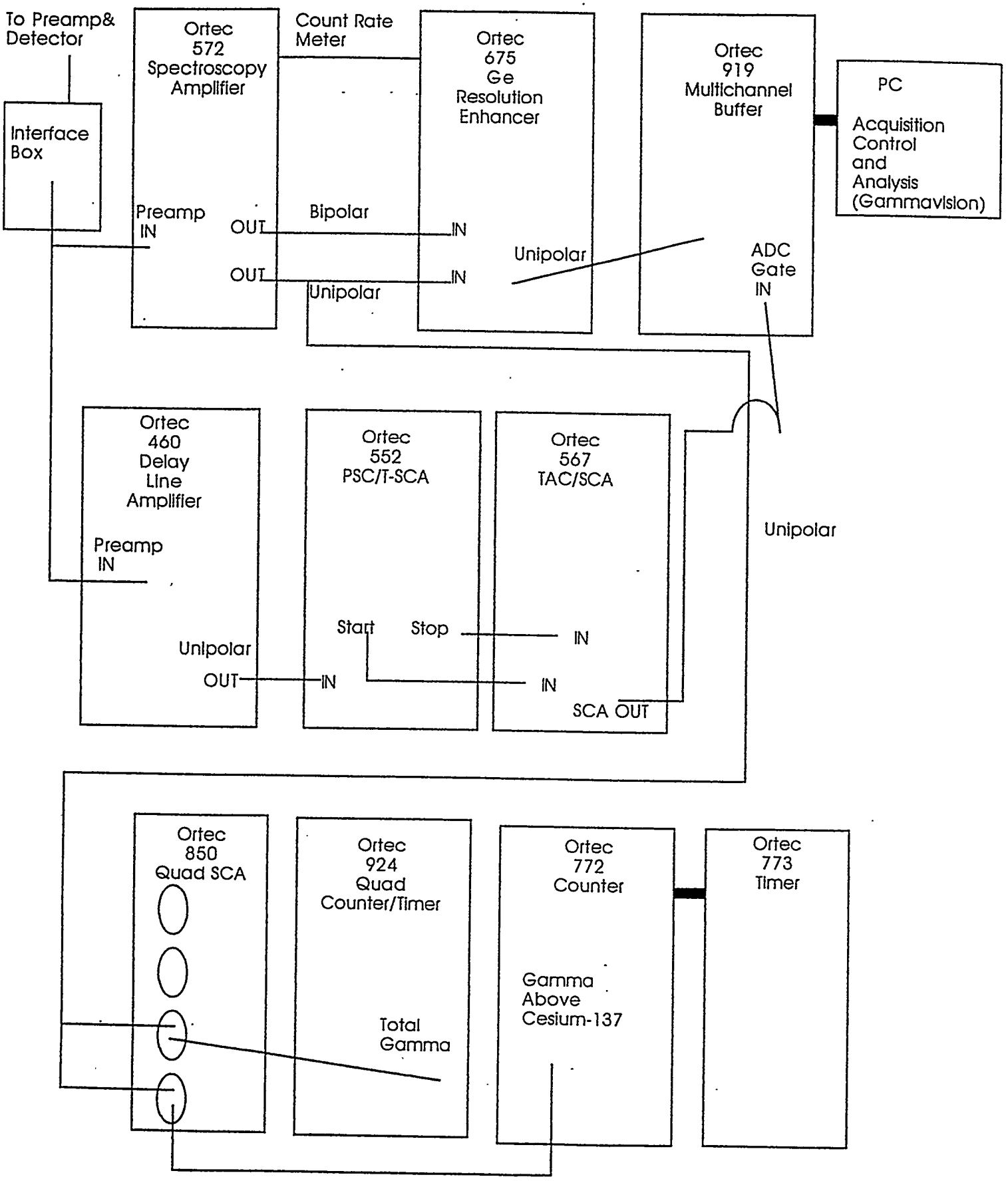
- SOFT SALT CAKE (DRY)
- SOFT SALT CAKE (WET)
- HARD SALT CAKE (DRY)
- HARD SALT CAKE (WET)
- HARD SLUDGE
- SOFT SLUDGE
- LIQUID
- WASH WATER
- SAMPLE FAILURE
- EMPTY SAMPLER
- HARD OBJECT / TANK BOTTOM
- SEGMENT NOT TAKEN

Profiles below indicate primary constituent in sampler and are intended to reflect tank waste conditions.
In some instances, extrusion information may be adjusted knowing other key information such as previous water additions.

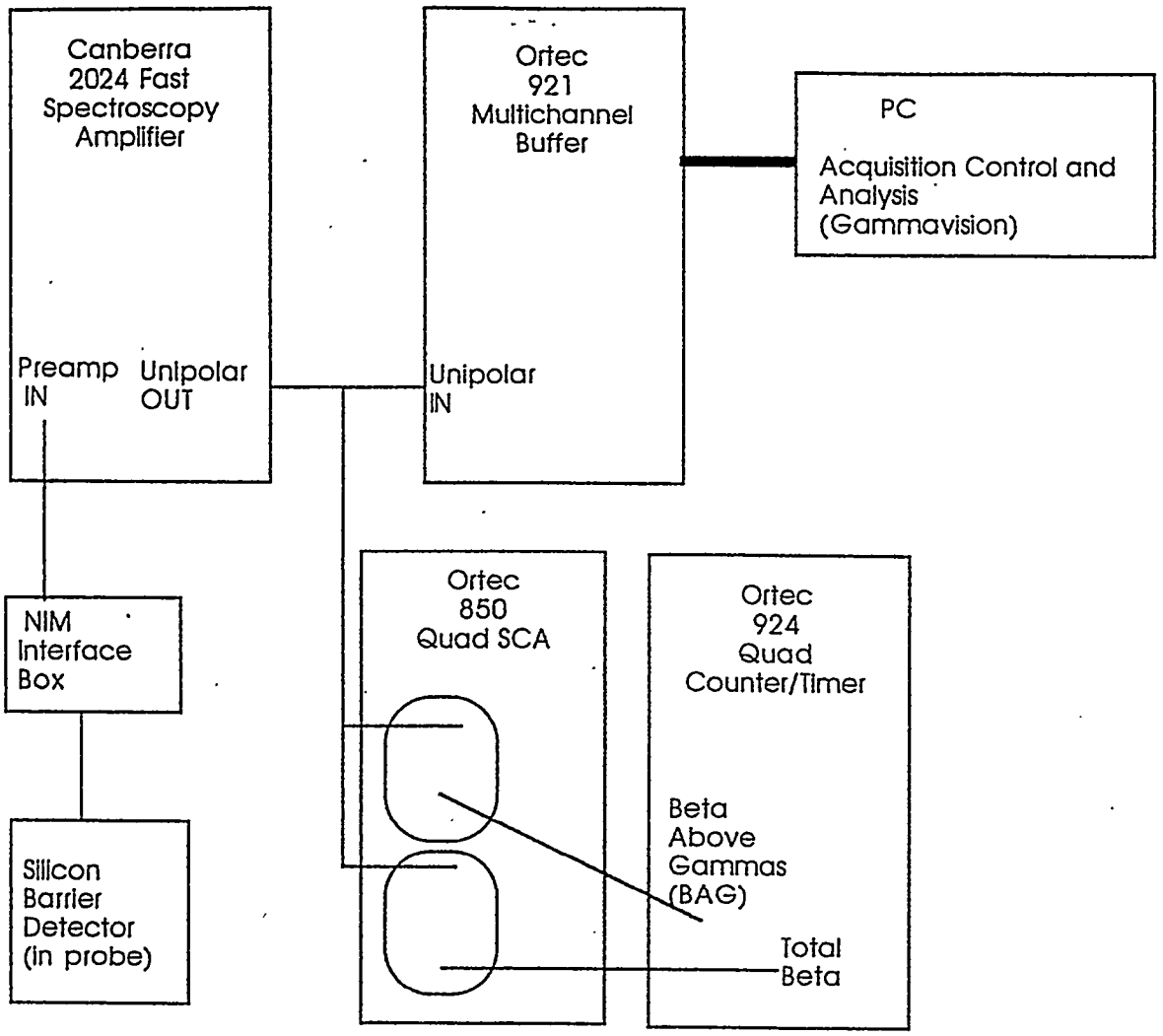


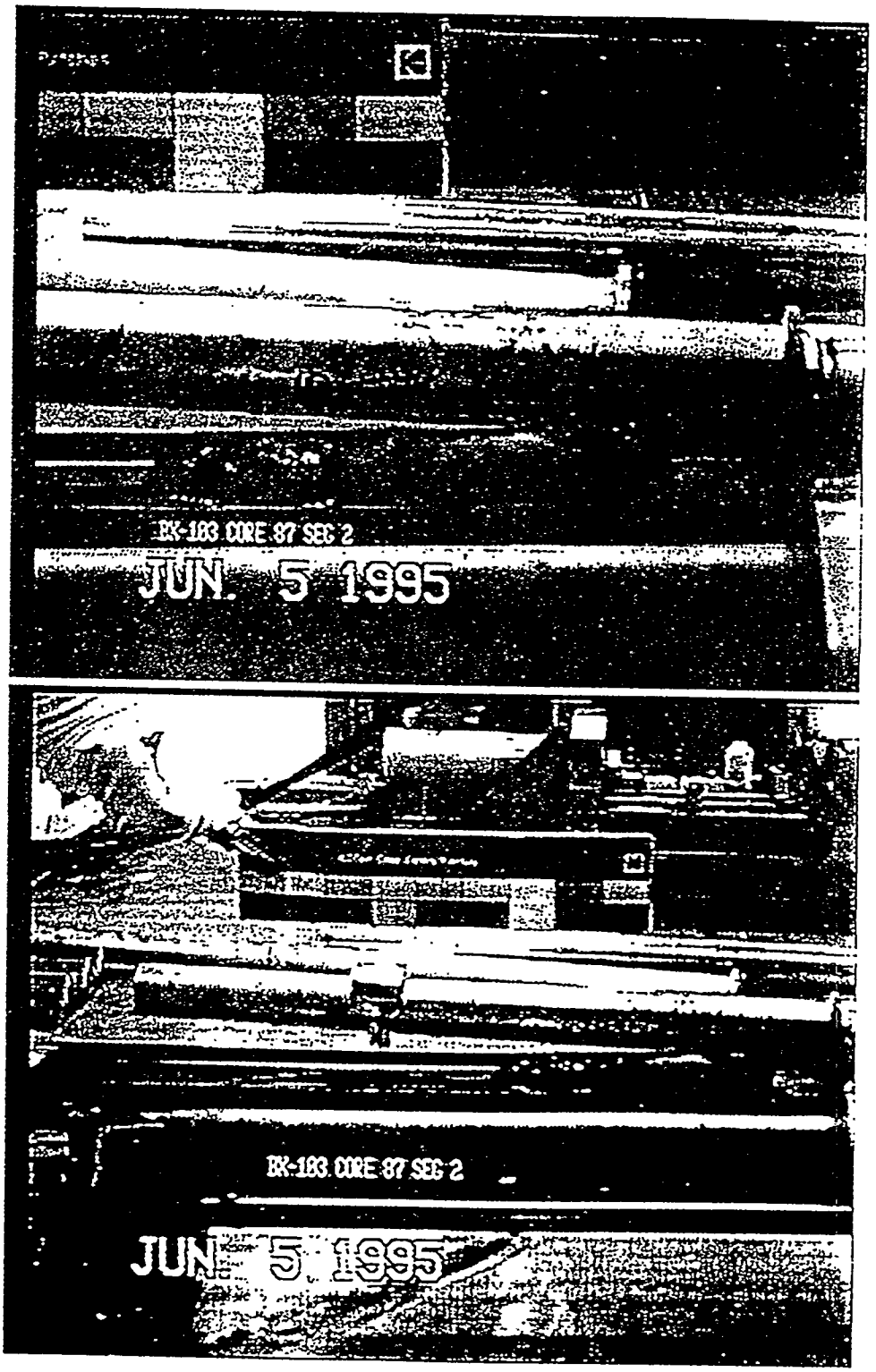
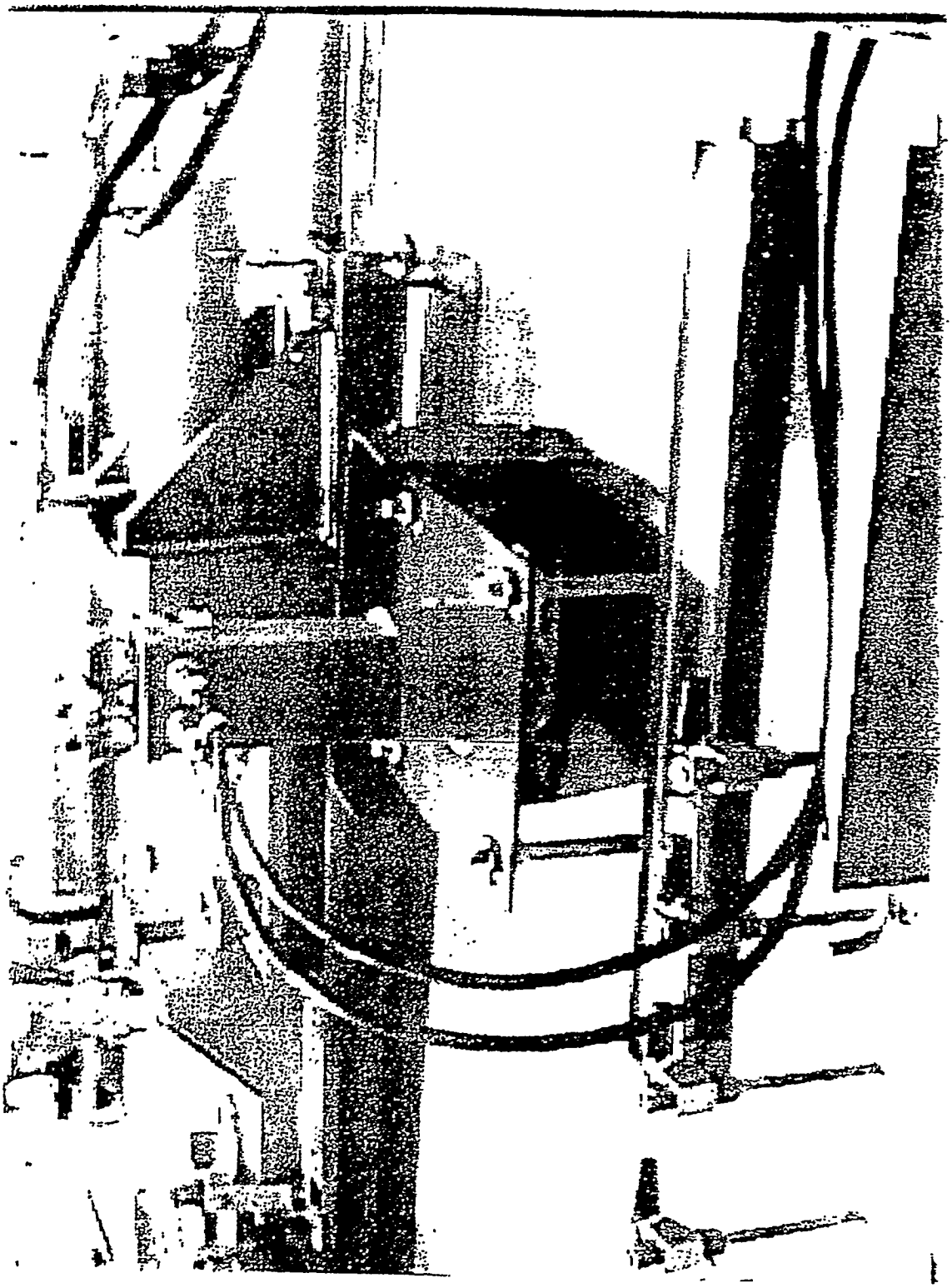


Gamma Spectroscopy System As Built



Beta Detection Electronics as Built

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