

Cerenkov Counter for In-Situ Groundwater Monitoring of ^{90}Sr

Robert C. Runkle^{*}, Ronald L. Brodzinski, David V. Jordan, John S. Hartman, Walter K. Hensley, Melody A. Maynard, William A. Sliger, John E. Smart and Lindsay C. Todd

Pacific Northwest National Laboratory, 902 Battelle Boulevard, P.O. Box 999, Richland, WA, 99352

* Author to whom correspondence should be addressed. E-mail: robert.runkle@pnl.gov

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Abstract: Groundwater contamination from ^{90}Sr is an environmental challenge posed to present and former nuclear weapons related sites. Traditional methods of extracting groundwater samples and performing laboratory analyses are expensive, time-consuming and induce significant disposal challenges. The authors present here a prototype counter capable of measuring ^{90}Sr groundwater concentrations in-situ at or below the drinking water limit of 8 pCi/liter. The ^{90}Y daughter of ^{90}Sr produces high-energy electrons, which can create Cerenkov light. Photomultiplier tubes convert the Cerenkov light into an electronic pulse, which then undergoes signal processing with standard electronics. Strontium-90 concentrations near the drinking water limit can be measured in a matter of hours if it is in secular equilibrium with the ^{90}Y daughter. The prototype counter is compact, can be deployed in an American Standard 6-inch, well while operated by a single person, and transmits the results to a central monitoring location.

Keywords: strontium-90, groundwater monitoring, Cerenkov light.

Introduction

The radionuclide ^{90}Sr is a common fission product with a half-life of 28.78 years. Large quantities of ^{90}Sr exist in effluent produced by fuel reprocessing and reactor operation. Because it concentrates in the skeletal system of humans, it presents an environmental challenge to the Department of Energy (DOE) at nuclear weapons related sites such as: Savannah River, South Carolina; Oak Ridge, Tennessee; Los Alamos, New Mexico; and Hanford, Washington. This issue is especially pertinent to the Hanford site where plumes of ^{90}Sr effluent present in ground water enter the Columbia River.

Groundwater concentrations are presently monitored in areas where known ^{90}Sr plumes reside. The goal is to ensure that the concentrations entering the Columbia River remain well below the free-release limit of 8 pCi/liter and to assess the performance of remediation efforts. Current monitoring practices are time-consuming and expensive. They require extracting down well samples, performing laboratory analysis, and waste disposal.

The authors present here a significantly more efficient method for monitoring ^{90}Sr groundwater concentrations, which can be performed in-situ and on a daily basis. This method also provides immediate results and creates no disposal problems. The prototype counter described below is an improvement over previous designs [1] that measure ^{90}Sr concentrations via the Cerenkov light produced from beta-decays.

Detector concept

Strontium-90 decays via beta-decay and emits an electron with a maximum energy of 546 keV. The daughter nucleus, ^{90}Y , has a half-life of only 64.10 hours and also undergoes beta-decay with a maximum electron energy of 2282 keV. The decay of ^{90}Y produces the stable isotope ^{90}Zr . Figure 1 shows the relative intensity of the emitted electrons as a function of energy. Note that the majority of ^{90}Y decays have electron energies above the ^{90}Sr endpoint.

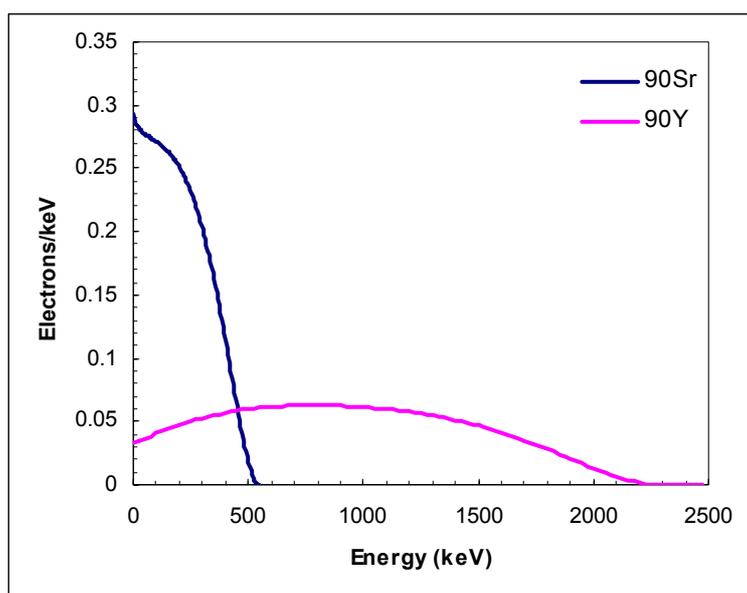


Figure 1. Graph of relative intensity for electrons emitted by ^{90}Sr and ^{90}Y .

High energy charged particles (electrons above ~ 250 keV in water) traveling through optically transparent materials with an index of refraction greater than unity produce Cerenkov light. The particle's momentum restricts this light emission to a cone centered along the direction of the particle's path. The Cerenkov light that reaches the photocathode of a photomultiplier tube (PMT) can be converted into an electronic signal. The pulse height of this signal is proportional to the energy of the charged particle.

Since water is transparent to Cerenkov light and can easily be confined to a known volume, measurement of Cerenkov light provides a simple technique for measuring high-energy beta decays occurring in a groundwater sample. The majority of ^{90}Sr decays will not produce Cerenkov light since their energy will be below the ~ 250 -keV threshold. In contrast, ^{90}Y is a prolific producer of Cerenkov light, due to its higher mean beta-decay energy. An unambiguous measurement of decays from ^{90}Y results from only counting events recorded above the ^{90}Sr endpoint. A sample's ^{90}Y activity is a function of the initial ^{90}Sr and ^{90}Y activities, $A_{^{90}\text{Sr}}^o$ and $A_{^{90}\text{Y}}^o$, and the decay constants, λ , for each nuclei:

$$A_{^{90}\text{Y}} = A_{^{90}\text{Y}}^o \cdot e^{-\lambda_{^{90}\text{Y}}t} + \frac{\lambda_{^{90}\text{Y}}A_{^{90}\text{Sr}}^o}{\lambda_{^{90}\text{Y}} - \lambda_{^{90}\text{Sr}}} \cdot \left(e^{-\lambda_{^{90}\text{Sr}}t} - e^{-\lambda_{^{90}\text{Y}}t} \right) \quad (1)$$

The ^{90}Sr concentration of samples in secular equilibrium can be deduced from a single measurement of the ^{90}Y activity, since secular equilibrium implies a ^{90}Sr to ^{90}Y initial activity ratio ($A_{^{90}\text{Sr}}^o : A_{^{90}\text{Y}}^o$) of unity, which is temporally constant. But there is no assurance that samples collected in the field exist in secular equilibrium, since the chemical interactions in soil and other permeable barriers may affect the movement/retention of strontium and yttrium differently. Samples not in secular equilibrium may have an initial activity ratio of Sr/Y greater or less than one. Those with a ratio greater than unity correspond to environments where ^{90}Sr more freely enters the groundwater, or, alternatively, where the ^{90}Y would be preferentially removed from the groundwater by the soil. However, because soil generally has a larger affinity for ^{90}Sr , an initial activity ratio less than unity is more common. In this case, the ^{90}Y activity will decrease with time and asymptotically approach the point of secular equilibrium, where the two activities are equal.

Figure 2 shows the behavior of the ^{90}Y activity for various initial activity ratios where the ^{90}Y activity exceeds the ^{90}Sr activity and has an initial value of unity (in arbitrary units). By securing the sample in the chemically stable environment of the counter, one ensures that ^{90}Sr and ^{90}Y are lost only via beta-decays. After measuring the ^{90}Y activity at two or more points in time, the data can be fit to the decay curves of Figure 2; the ^{90}Sr concentration can then be deduced.

Beta decay is not the only process that produces Cerenkov light; cosmic rays and terrestrial γ -rays may also contribute. Background from cosmic rays primarily results from high-energy (GeV) muons created in the upper atmosphere. These muons interact in the same way high-energy electrons do. Similarly, terrestrial γ -rays above approximately 410 keV can Compton scatter in water producing electrons of varying energies, which themselves then can create Cerenkov light. The principle sources of terrestrial radioactivity stem from nuclei in the ^{232}Th and ^{238}U decay chains along with ^{40}K , all of which are present in soil.

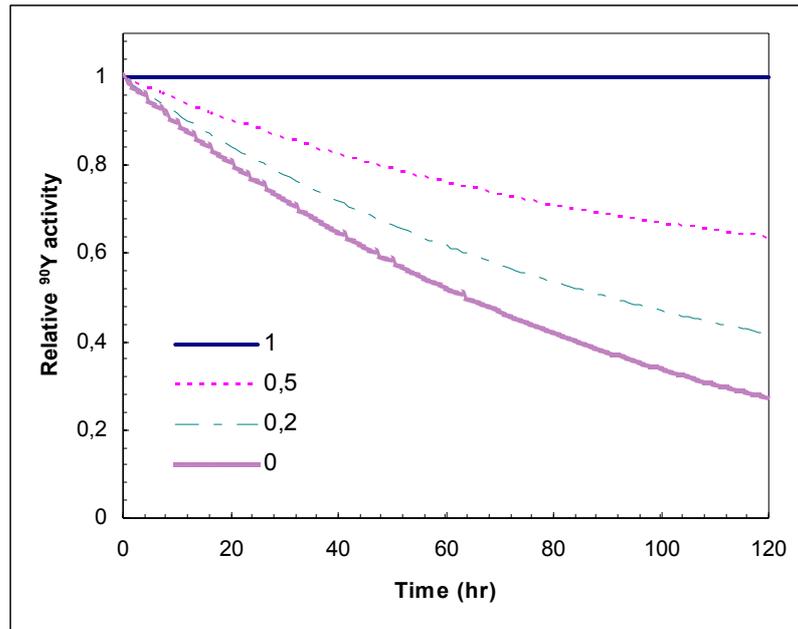


Figure 2. Graph of the relative ^{90}Y activity as a function of the initial activity ratio ($A_{^{90}\text{Sr}}^o : A_{^{90}\text{Y}}^o$). The four ratios plotted above are 1, 0.5, 0.2, and 0. The top line, corresponding to an initial activity ratio of unity, results from a sample initially existing in secular equilibrium. The remaining curves asymptotically approach secular equilibrium. The bottom curve, representing the case where no ^{90}Sr is present, asymptotically approaches zero and is simply the ^{90}Y decay curve with a 64.1 hour half-life. Not shown here are curves that would increase if the initial ^{90}Sr concentration exceeded that of ^{90}Y .

The contributions from these sources of background vary depending on the field environment. Bowyer, et al., [1] reported cosmic rays as the dominant source of background. However, for down-well applications, where the counter is surrounded by soil containing terrestrial radioactivity, γ -rays present the largest background.

Physical modeling and conceptual design

The chief constraints on the detector design presented here derive from two factors. First, it has to fit down-well, which in our specific application implies a diameter of less than 15 cm and a maximum total length of 1.8 m. Second, the counter must accurately measure concentrations down to 8 pCi/liter, the drinking water limit, in a reasonable amount of time, i.e. hours. This requires maximizing the counter's detection efficiency. With these general parameters in mind, the authors investigated various geometries using the simulation code GEANT4 [2] which modeled all aspects of the detector from the energy distribution of electrons produced during beta-decay to the photomultiplier tube response.

Figure 3 displays a typical simulation geometry in which the counter is immersed in a volume of soil. Model calculations varied the inner diameter and length of the water cavity within the limits mentioned above. The top and bottom end of the cavity were fit with quartz windows while the inside surface was lined with Teflon (95% reflectivity). Calculations included light attenuation and internal reflections. All beta-decays were produced at points randomly distributed throughout the water volume. Although the simulation results were only used in constraining the design parameters

discussed below, prior to counter construction their accuracy was partially validated by reproducing the detection efficiency of the Bowyer counter [1].

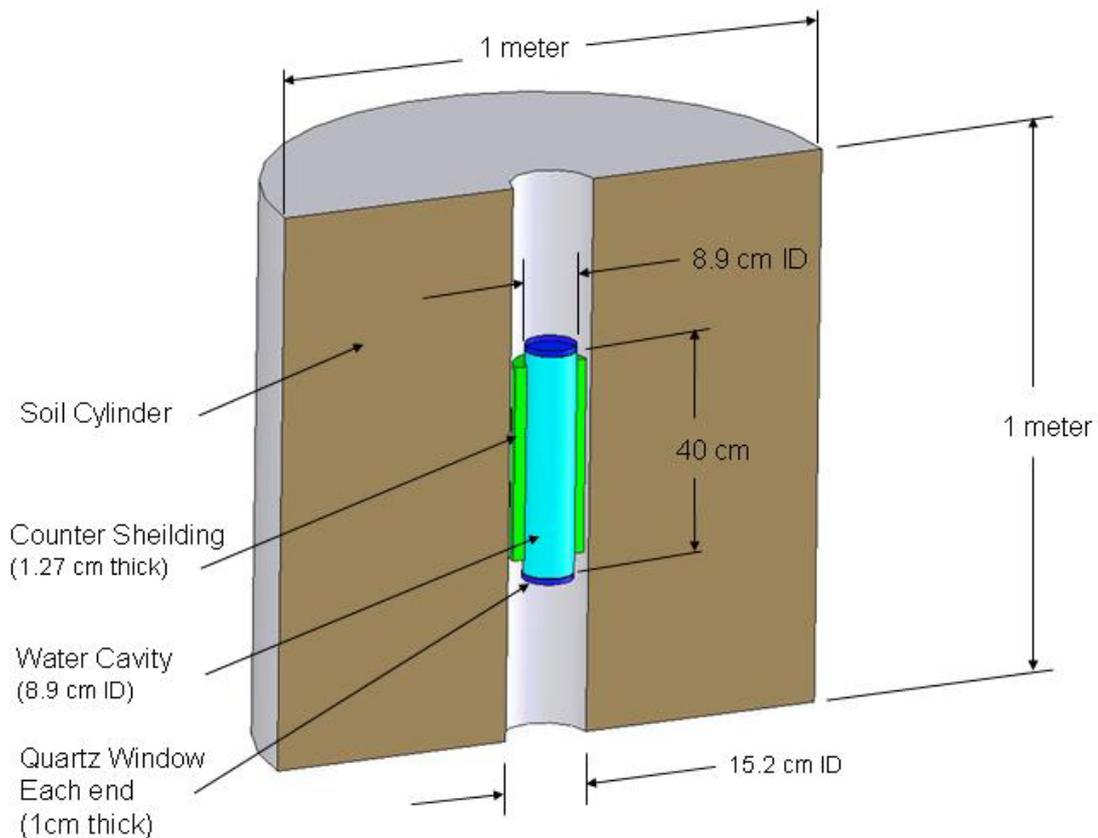


Figure 3. The simulation geometry includes a water cavity, shielding (active or passive), and surrounding soil. Essentially all of the gamma-ray flux entering the counter is accounted for with the 1-m right circular cylinder of soil shown here. Not shown in the figure is a disk above the counter from which trajectories of cosmic rays were calculated.

The most important design parameter influencing the detection efficiency, i.e. count rate, is the water cavity volume. For a fixed diameter, the count rate increases with the counter length due to the presence of more beta-decays. The relationship is asymptotic due to the attenuation of light traveling through large distances of water. A further complexity is the existence of dark current in PMTs. PMT dark currents create electronic noise and false signals. One technique for dramatically reducing this effect is to require a coincidence between two PMTs. Since the propagation of Cerenkov light is restricted to a cone in the direction of the electron's momentum, requiring a coincidence between multiple PMTs installed on one of the counter's ends is one option for reducing noise, i.e. single-ended coincidence. A second option is to employ one PMT on each end surrounded by a Teflon reflecting ring.

As shown in Figure 4, light that is emitted towards one end of the tube will reflect and, if not attenuated, create a coincidence at the opposite end, i.e. two-end coincidence.

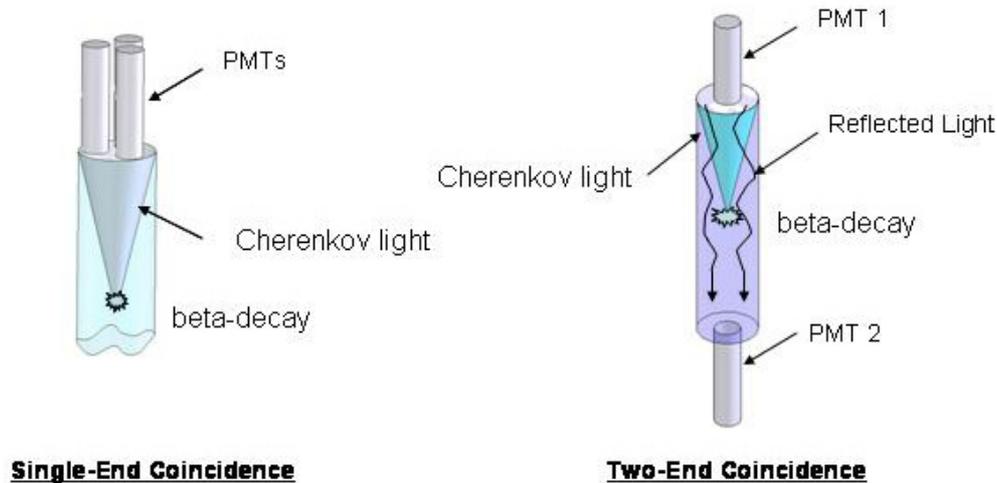


Figure 4. Two possible coincidence techniques that serve to reject PMT dark current events.

Figure 5 compares the signal rate for various PMT configurations as a function of water cavity length and coincidence requirements. Single-ended coincidence schemes have an asymptotic behavior while configurations with an opposite-end coincidence requirement have an optimal length. This peaked behavior results from a balance of having more signal events in a longer counter with the attenuation of the reflected light over the length of the counter.

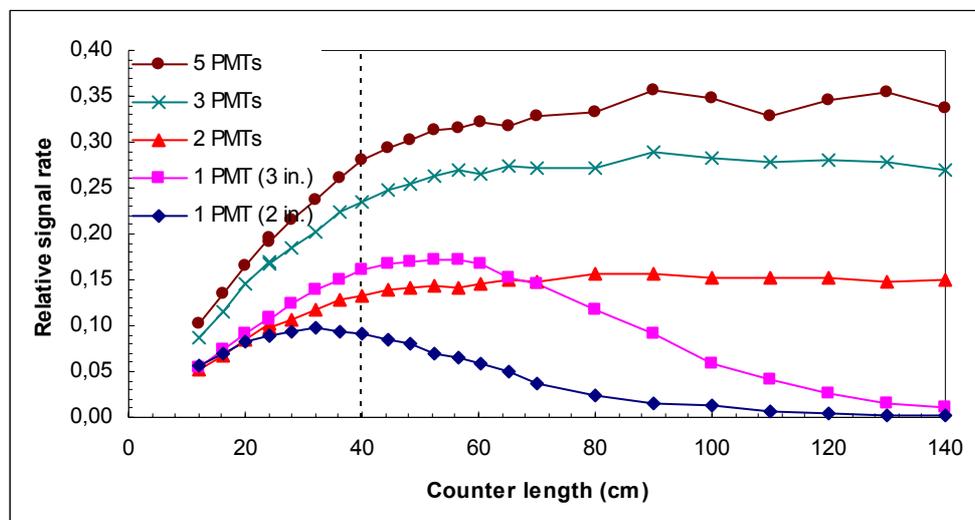


Figure 5. Graph of the relative event rate in the water cavity as a function of counter length and number of PMTs on each end. Configurations with more than one PMT per end have single-end coincidence requirements while those with only one PMT have a two-end requirement.

The second crucial design parameter is the background shielding configuration. Both terrestrial gamma rays and cosmic rays were included in the background simulation. The soil served as a generator of the γ -ray background, and the model assumed soil activity concentrations of 0.7 pCi/g for ^{232}Th , 0.4 pCi/g for ^{238}U , and 10 pCi/g for ^{40}K [3]. The cosmic-ray generator consisted of muons with initial positions uniformly distributed on a disk-shaped surface above the counter with a momentum distribution given by Kremer, et al., [4] and an angular distribution of $\cos^2(\theta)$, where θ is the angle

normal to the soil. The resulting event rates in an 8.9-cm diameter by 40-cm long cylinder with no shielding were 1800 Hz for gamma rays and 5 Hz for cosmic rays. This result focused the design on eliminating the terrestrial γ -ray background.

To this end, the authors considered both active and passive configurations for background reduction. The goal of active shielding is to detect the penetrating radiations so that coincident signals in the Cerenkov counter can be vetoed and not misidentified as beta-decay events; passive configurations attempt to absorb penetrating radiations before they enter the counter. The active method modeled here was a veto shield surrounding the sample cylinder composed of BGO, NaI(Tl) or CsI(Na). Both gamma rays and cosmic rays have a high probability of depositing some energy in these materials, especially BGO. The authors also considered passive shields of lead and tungsten, which absorb gamma rays from the surrounding soil but have little impact on cosmic rays because of their enormous energy. Another background reduction strategy is to reject cosmic-ray events depositing large energies. An upper-level pulse height cut applied to the PMT signal above the maximum ^{90}Y energy greatly assists in reducing the cosmic-ray background rate. The count rates for a 1.27 cm thick annulus surrounding the water cavity from each approach are displayed in Figure 6. The active veto shields are significantly more effective in rejecting background radiation.

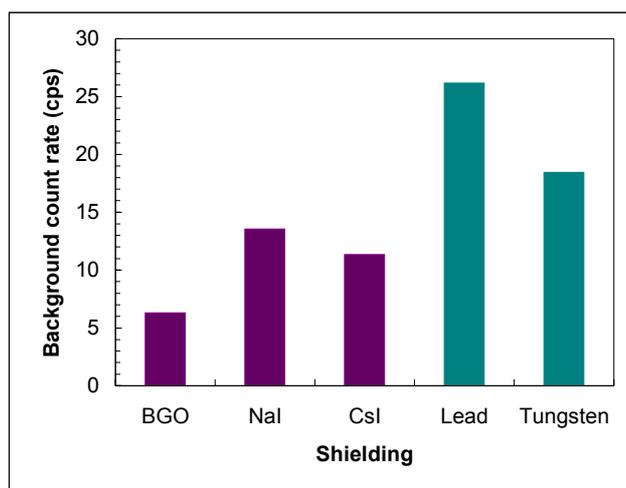


Figure 6. Background count rates for a cylindrical water cavity 40 cm long and 8.9 cm in diameter with 4 PMTs at each end operated in a single-ended coincidence mode. In each case, the shield is an annulus 1.27 cm in diameter. The three active shields considered were the scintillators BGO, NaI(Tl) and CsI(Na); lead and tungsten are passive shields.

Counter design

Based on the simulation results discussed above and costs associated with design, materials, and electronics, the decision to build a prototype counter with a single PMT on each end and passive tungsten shielding was made. The prototype counter design consists of a cylindrical water cavity, 8.9 cm in diameter and 40 cm long, defined by a tungsten annulus of thickness 2.1 cm, as shown in Figure 7. Two quartz windows provide a water tight and transparent seal for 5.08-cm diameter PMTs on each end. The 40-cm cavity length is optimal for this PMT configuration. The inside of the tungsten

cavity is coated with a Teflon reflector including the annular area adjacent to the PMTs. A stainless-steel housing encloses the tungsten annulus, PMTs, and space for a water pump, vent line, and electronics.

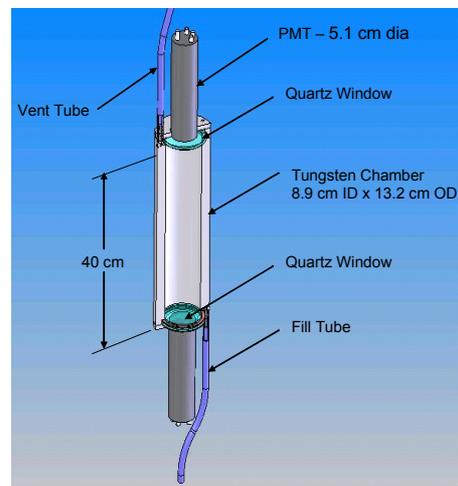


Figure 7. Schematic of the ^{90}Sr final design which includes a passive tungsten shield and two 5.1-cm diameter PMTs.

In the prototype design reported here, all signal processing occurs at the surface of the well is using standard NIM electronics. High voltage enters, and signals exit, the counter through water-tight connections on its top. The signal processing electronics shown in Figure 8 have two principal features: 1) a lower level discriminator on each PMT and a coincidence requirement between both PMTs to prevent triggering on electronic noise; and 2) an upper level discriminator on the sum of both PMT signals to reject cosmic-ray events above the maximum ^{90}Y electron energy. Signals remaining after applying these criteria are counted as potential ^{90}Y decays and sent to a laptop computer.

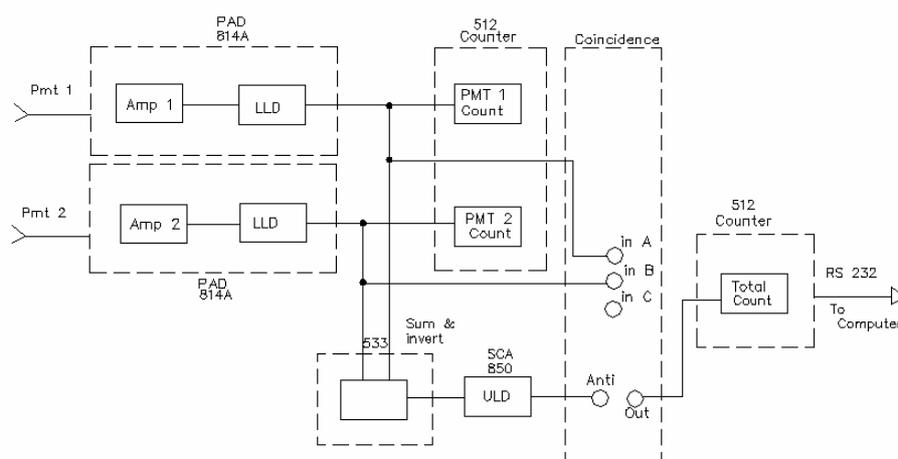


Figure 8. The signal processing electronics, presently located above ground, incorporate the Canberra modules noted above. The two essential criteria are the coincidence requirement between each phototube and an anti-coincidence veto if the signal exceeds an upper level discriminator.

The authors tested this counter with samples from a Hanford site well whose ^{90}Sr concentrations were ~ 1000 pCi/liter, and which are in secular equilibrium. The authors counted the samples in an above ground laboratory environment where background from soil was reduced relative to down well operation. Figure 9 displays a pulse height spectrum for this sample. The region to the right of the vertical dashed line corresponds to the region above the lower level discriminator of Figure 8 while the right end of the graph marks the upper level cosmic-ray discriminator.

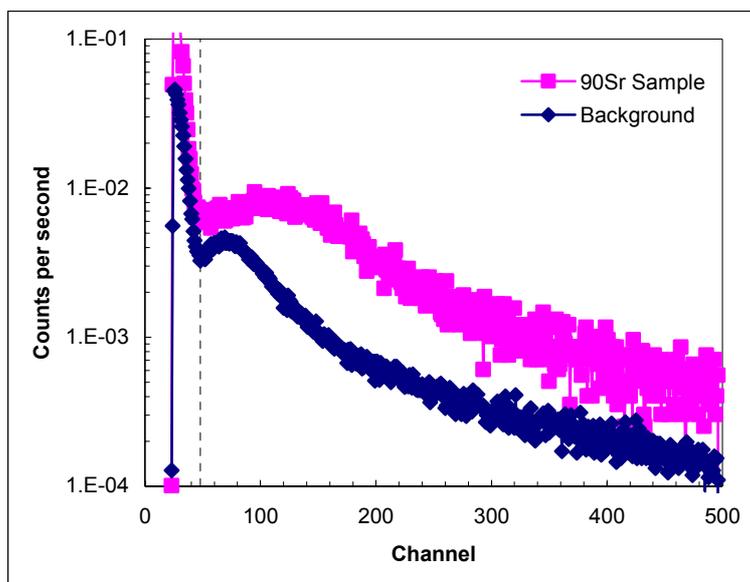


Figure 9. Pulse height spectrum for PMTs in two-end coincidence mode. The pulse height represents the sum of both PMTs.

Field operation

The ultimate use of this counter is for unmanned field measurements. The prototype presented here can be deployed from the back of a pickup truck and lowered into a well. (Future incarnations will contain all signal processing inside the stainless steel shell located down-well.) In either case, a power supply and transmitter located above ground feed the results via cellular communications to a central monitoring station. Water samples will be pumped into and out of the counter through an in-counter pump and particulate filter.

Conclusions

The authors constructed and calibrated a prototype counter for monitoring ^{90}Sr contamination in ground water. It consists of a cylindrical water cavity, 8.9 cm in diameter and 40 cm long. Groundwater samples with ^{90}Sr concentrations down to the drinking water limit of 8 pCi/liter can be analyzed on a daily basis. The detector is a field-deployable device and is equipped with wireless communications for remote monitoring.

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