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ABSTRACT

A cooperative program of measurements between Westinghouse Hanford Co. (WHC) and Oak Ridge National Laboratory (ORNL) have been performed to develop and test a high efficiency neutron detector for use in detecting the presence of fissionable material in the nuclear waste storage tanks at Hanford, Washington. The detector system developed at ORNL was successfully tested in tank TX-118 at the 200 W area tank farm at Hanford on October 19, 1994. Prior to the measurements in tank TX-118, the system was tested at the Radiation Calibration (RADCAL) facility at ORNL. The successful operation of the detection system in tank TX-118 will allow measurements to detect the fissionable material in the Hanford storage tanks to proceed as planned.

INTRODUCTION

A program of measurements and calculations to develop a method of measuring the fissionable material content of the large waste storage tanks at the Hanford site is described in this report. These tanks contain radioactive waste from the processing of irradiated fuel elements from the Pu producing nuclear reactors at the Hanford site. Time correlation and noise analysis techniques, similar to those developed for and utilized in the Nuclear Weapons Identification System (NWIS)[1], at the Y-12 Plant in Oak Ridge, will be used at the Hanford site. Both "passive" techniques to detect the neutrons emitted spontaneously from the waste in the tank, and "active" techniques using AmBe and ^{252}Cf neutron sources to induce fissions will be used. This work is divided into 3 major tasks: 1) development of high sensitivity neutron detectors that can selectively count only neutrons in the high γ-radiation fields in the tanks, 2) Monte Carlo neutron transport calculations using both the KENO[2] and MCNP[3] codes to plan and analyze the measurements, and 3) the measurement of time correlated neutrons by time and frequency analysis to distinguish spontaneous fission from (α,n) sources inside the tanks. This report describes the development of the detector and its testing in radiation fields at the RADCAL facility at ORNL and in tank TX-118 at the 200 W area at WHC.

PREVIOUS WHC MEASUREMENTS

Each of the waste tanks at Hanford has a Liquid Observation Well (LOW) through which measurements on the material in the tank can be made. A LOW is a fiberglass pipe which is sealed from the contents of the tank. It extends vertically from above ground to within a few inches of the bottom of the tank and is offset several feet from the center of the tank. WHC has used a BF_3 proportional counter which was 3.8 cm in diameter by 10 cm long, having a fill pressure of 1/3 atmosphere, with a sensitivity of approximately 8 counts per unit of thermal neutron flux to obtain the vertical profile of the spontaneous neutron flux inside the tanks. Periodically, the same detector has been used in conjunction with an AmBe neutron source to monitor the liquid level in the tanks.

CHOICE OF TANK TX-118

A vertical scan made with the WHC BF_3 detector inside the LOW of tank TX-118 showed that the background neutron flux was much higher than was typical for the other tanks at Hanford. The neutron flux was fairly constant at approximately 10 counts/sec between approximately 8 feet and 32 feet from the bottom of the tank and fell to zero both below and above this range. The neutron count rate profile provided by WHC is shown in Fig. 1. Because of the anomalously high count rate this tank was chosen by WHC for the first ORNL measurements. Measurements by WHC personnel also showed the ^{137}Cs γ-ray exposure rate in the tank to vary from approximately 30-60 R/hr.

ADVANTAGE OF CORRELATION MEASUREMENTS

Both (α,n) reactions (driven by the α-decay of the actinides in the tank) and spontaneous fission (from, for example, ^{240}Pu) are possible sources of the neutron flux in the tank. Using correlation measurements similar to those in use at the Oak Ridge, Tennessee Y-12 Plant to identify fissile material[1], it may be possible to measure whether the neutrons in tank TX-118 arise from (α,n) reactions or spontaneous fission. If the neutrons are a result of spontaneous fission then several neutrons are emitted from each fission; hence, these neutrons are correlated with one another. Therefore, it is in principle possible to measure the amount of correlation to determine estimates of the spontaneous fission rate, and hence the amount of fissionable material in the tank. By using additional neutron sources, like ^{252}Cf or AmBe inserted into the LOW, correlated neutrons will be produced by any induced fission in the materials in the tank. Because of the geometry involved (very large source area and relatively small available volume for detectors) and the intense gamma radiation background in the tank, such a correlation measurement is expected to be difficult. The main goal is to obtain the highest possible detection efficiency at an acceptable signal-to-noise ratio.

NEED FOR A HIGH EFFICIENCY DETECTOR

Detectors more efficient than the one used by WHC will be needed to perform the correlation measurements. This is because the method requires the use of at least two detectors and the correlated counting rate scales approximately as the square of the detector efficiency. Assuming spontaneous fission to be the neutron source, MCNP calculations made at ORNL indicate that the Hanford BF_3 detector has an efficiency of approximately 9.8×10^{-7} for the emitted fission neutrons in tank TX-118 and that the total neutron emission rate from spontaneous fission was approximately 1.0×10^{7} neutrons/s. Therefore, the expected coincidence rate using two of these detectors is approximately 3.8×10^{-6} s^{-1} (assuming 2.5 neutrons are emitted per fission). Because of this very small coincidence rate, passive correlation measurements are impractical with detectors of this type. This rate should be increased substantially to perform measurements in a reasonable time. To determine the expected efficiencies and counting rates, MCNP calculations were performed with several different detectors. The calculations indicated that it should be possible to increase the detector efficiency by as much as a factor of 25 by using readily available ^{3}He proportional counters. Furthermore, because the detectors absorb most of the low-energy neutrons, the efficiency does not simply scale with detector volume and gas pressure. For example, for detectors which were 5.1 cm in diameter by 91 cm long, the MCNP calculations indicate that 1.5 and 0.75 ATM detectors would be 70% and 40%, respectively, as efficient as a 3 atm detector. A remaining question was whether these more efficient detectors would have adequate signal-to-noise ratio in the high-γ-background inside the tank.

APPROACH TAKEN TO INCREASE DETECTOR EFFICIENCY

To obtain better discrimination against γ-rays, CF_4 gas was added to the ^{3}He proportional counters. The CF_4 additive increases the drift velocity of the gas and localizes the volume where the ionization is produced. As a result, the ionization from the reaction products is produced in a shorter time and a shorter amplifier shaping time can be used. This decreases γ-ray pileup effects so that good γ-ray discrimination can be obtained at the same time as higher neutron sensitivity.

RADCAL MEASUREMENTS

To ascertain whether the more efficient detectors would work in the high-γ-background environment of tank TX-118, several detectors were tested in a calibrated γ-ray field at the RADCAL Facility at ORNL. Data supplied by WHC indicated that the highest γ-radiation levels in this tank were 60 R/hr inside the salt cake at the bottom of the tank, and about 30 R/hr above the salt cake. At RADCAL γ-ray fields up to 208 R/hr can be obtained at a distance of 12 cm from a calibrated ^{137}Cs source (the same γ-ray source as in TX-118). Lower radiation levels can be obtained by moving the detector away from the source. Because the source is collimated, the area covered by the γ-ray field depends on the distance from the source and the maximum dose is available over an area only 2 inches in diameter. The highest fields were obtained with the detector axis parallel to the γ-ray beam so that only one end of the detector was at the highest rate, and the exposure rate decreased rather steeply along with the length of the detector. On the other hand, at a exposure rate of 30 R/hr (obtained at 31.6 cm from the source) almost all of an 45.7-cm-long detector could be placed inside the beam when the detector axis was perpendicular to the beam. For these reasons, exposure rates greater than 30 R/hr were only approximate. As a check of the effect of the variation of the exposure rate along the length of the detector, tests were made with an 45.7-cm-long detector both parallel and perpendicular to the γ-ray beam at a distance of 31.6 cm from the source. There was no measurable difference between the pulse-height spectra taken under these two conditions.

Most of the measurements were made with ^3He proportional counters which were nominally 5.1 cm in diameter by 45.7 cm long. Three different combinations of filling gasses were used: 1) 98% ^3He - 2% CO_2 with a total pressure of 3 atm., 2) 74% ^3He - 26% CF_4 with a total pressure of 2.7 atm., and 3) 75% ^3He - 25% CF_4 with a total pressure of 1 atm. These proportional counters are designated as detectors 1, 2, and 3, respectively in the following discussion. In addition, the detector high voltage and amplifier shaping time constant were varied to optimize the discrimination against γ-rays.

The RADCAL measurements were performed using a moderated PuBe neutron source at a distance of approximately 1 m from the detector. This distance was chosen to obtain approximately the same neutron counting rate as expected inside tank TX-118 at Hanford. To overcome the background from the pileup of the pulses due to the intense γ-ray field it was necessary to operate the detectors at much shorter shaping time constants and lower voltages than are typically used when counting neutrons without an intense γ-ray background. For example, the best results were obtained at a high voltage of 1640 V and amplifier shaping time constant of 0.25 μs for detector 1, 2250 V and 0.25 μs for detector 2, and 1300 V and 0.5 μs for detector 3.

Detector 2 was found to be best overall. Approximately 2/3 of the neutron signals were above the γ-ray background. The absolute neutron efficiency of detector 2 (above the γ-ray background) was about 50% greater than detector 3. The absolute neutron efficiency above the γ-ray background of detector 1 was approximately the same as detector 2. However, to avoid being overwhelmed by γ-ray pileup, it was necessary to operate detector 1 under conditions in which there was no clear neutron peak in the pulse-height spectrum. In contrast, both detectors 2 and 3 gave a clear neutron peak above the γ-ray background. The absence of a neutron peak in the spectrum for detector 1 would make it difficult to monitor the stability of the detector during actual measurements at Hanford and to verify the counting of neutrons. The effects on the pulse-height spectrum of detector 1 of increasing the γ-ray exposure are shown in Fig. 2 where spectra for exposure rates of 0, 30, 60, and 208 R/hr are shown. As shown in Figs. 3 and 4, the addition of CF_4 gas to the ^3He detectors improved the pulse-height distribution substantially so that a clear neutron peak was observable above the γ-ray noise.

HANFORD MEASUREMENTS

On October 19, 1994, detectors 2 and 3 were assembled in an apparatus (called the detector jig) and lowered into the LOW in tank TX-118 at Hanford. The detector jig consisted of a stainless-steel pipe which was 7.0 cm in diameter by 3.1 m in length, which held both detectors and their preamplifiers, and a DC-to-DC converter to supply the detectors with high voltage from input low voltage. A multiconductor cable was used to supply the necessary voltages to the preamplifiers and the detectors as well as to obtain the output signals from the detectors. A sketch of the TX-118 LOW detector configuration is given in Fig. 5. A vertical scan of the tank from 11 to 39 ft from the bottom was made inside the LOW using detector 3. The pulse-height spectra obtained at 8 positions are shown in Fig. 3. These spectra show a clear neutron peak above the pileup noise from the γ-rays. As shown in Fig. 6, the counting rate in the neutron peak was approximately constant with the position of the detector inside the LOW, except for the highest position where it fell dramatically. This behavior is in agreement with the earlier WHC data shown in Fig. 1. The counting rate was approximately 80 counts/s, or about 8 times larger than with the WHC BF_3 detector. This rate is in reasonable agreement with what was expected from the results of the MCNP calculations and the RADCAL measurements.

In Fig. 4 the pulse-height spectrum measured with detector 3 at RADCAL, at a exposure rate of 30 R/hr, is compared to the spectrum measured in tank TX-118. These spectra show that the measurements at RADCAL provided a good prediction of the performance of the detector in tank TX-118. Pulse-height spectra were not obtained with detector 2 because the electronic noise was too high. This noise was apparently caused by the generator which supplied the electrical power for all of the equipment. However, the signals from detector 2 were observed on an oscilloscope and appeared to be as expected except for the electronic noise. Hence we are confident that if cleaner electrical power is available that detector 2 will work as expected, which should allow us to obtain a 50% higher counting rate than with detector 3.

Based on the results of the measurements at RADCAL and at WHC, a 152-cm-long detector of the same diameter as the prototype detectors is practical. The 152-cm-long detector having the same fill gas as detector 2 would have a neutron counting rate above the γ-ray pileup about a factor of 42 times larger than the WHC BF_3 detector. This should increase the passive coincidence neutron counting rate in a correlation measurement between a pair of such detectors by a factor of over 1700, compared to using two of the WHC BF_3 detectors, if the neutron flux is due to spontaneous fission. Although the correlated neutron counting rate still would be fairly low, a passive measurement to ascertain the presence of fission neutrons should be possible in less than 1 day. Active measurements with an AmBe or ^{252}Cf neutron source could require less time.

OTHER OBJECTIVE OF THE TESTS

The test had the additional objective of obtaining actual practical experience of putting a detector into the LOW of tank TX-118. Based on these tests it is not practical to make any significant changes to the detection system at the tank farm site. Modifications to our hardware for the correlation tests will be incorporated to facilitate ease of changing detectors and adding and removing sources. For the correlation measurements, the detectors will be installed and tested in the tubes to be inserted into the LOW prior to arriving at WHC. Because of the electrical noise problem caused by the generator, electrical power should be supplied from another source for the next measurements. Check out and calibration of the total system will be performed at ORNL to verify proper operation prior to use at WHC.

CONCLUSIONS

These measurements showed that the ORNL developed detector operated successfully in tank TX-118 at the WHC tank farm. The performance of the detector was close to that expected from measurements and calculations made at ORNL. This gives us confidence that we will be able to field an even larger detector having a factor of 42 greater efficiency than the WHC BF_3 detectors. The higher efficiency detectors will be essential for determining the amount of the fissile material in the tanks through correlation measurements. The lessons learned by actually installing and operating a detector will be incorporated into the design of the detection modules for the correlation measurements in TX-118 which is anticipated to be the next step in this program.

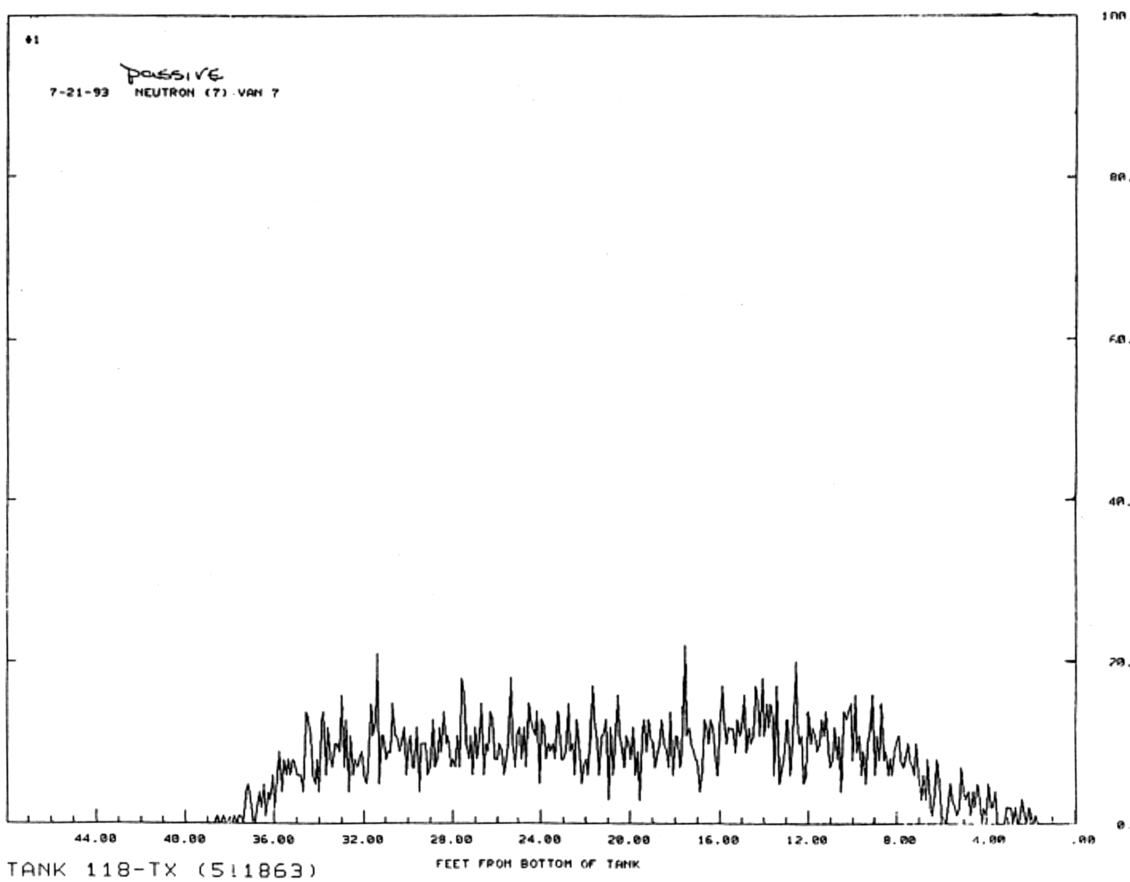

Fig. 1. Neutron counting rate versus distance of the detector from the bottom of tank TX-118. These data were provided by WHC and were made with a BF_3 detector which was 3.8 cm in diameter by 10 cm long and had a pressure of 1/3 atmosphere.

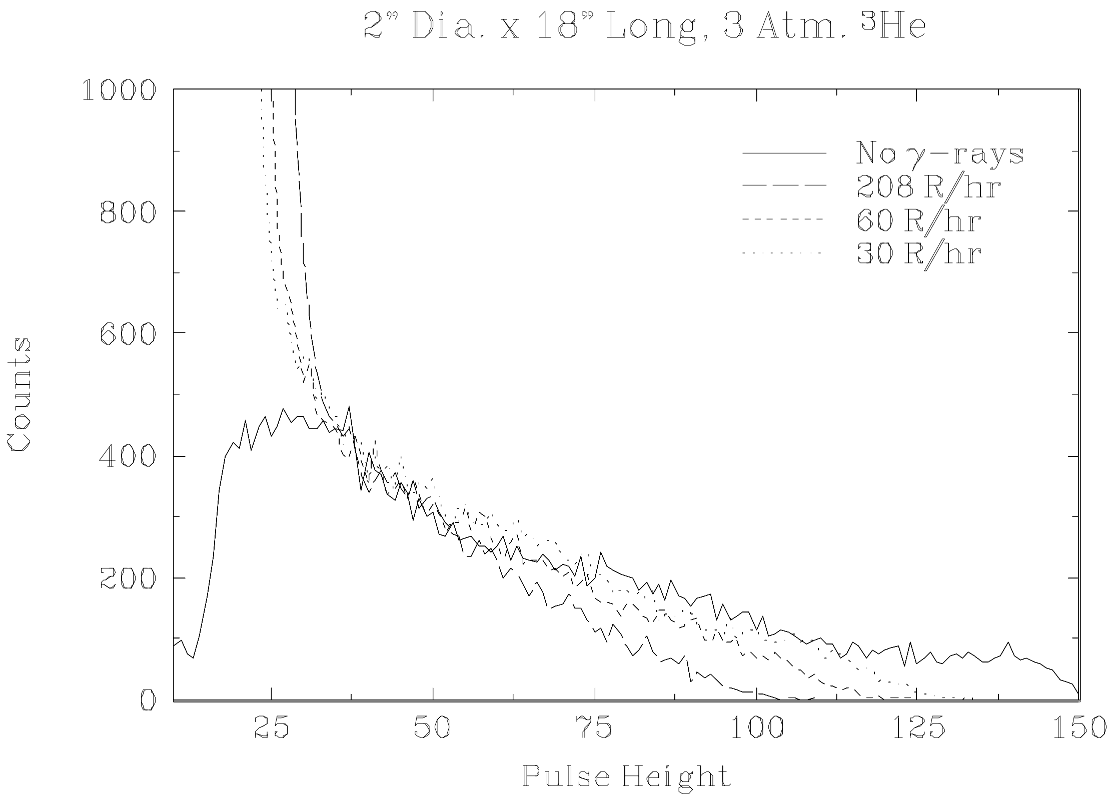

Fig. 2 Measurements made with ORNL ^{3}He detector 1 (5.1 cm diameter by 45.7 cm long, total pressure of 3 atm., 98% ^{3}He - 2% CO_2) at RADCAL. These data show the effect of an increasing γ-ray field on this detector. Spectra taken with an improved detector are shown in Figs. 3 and 4.

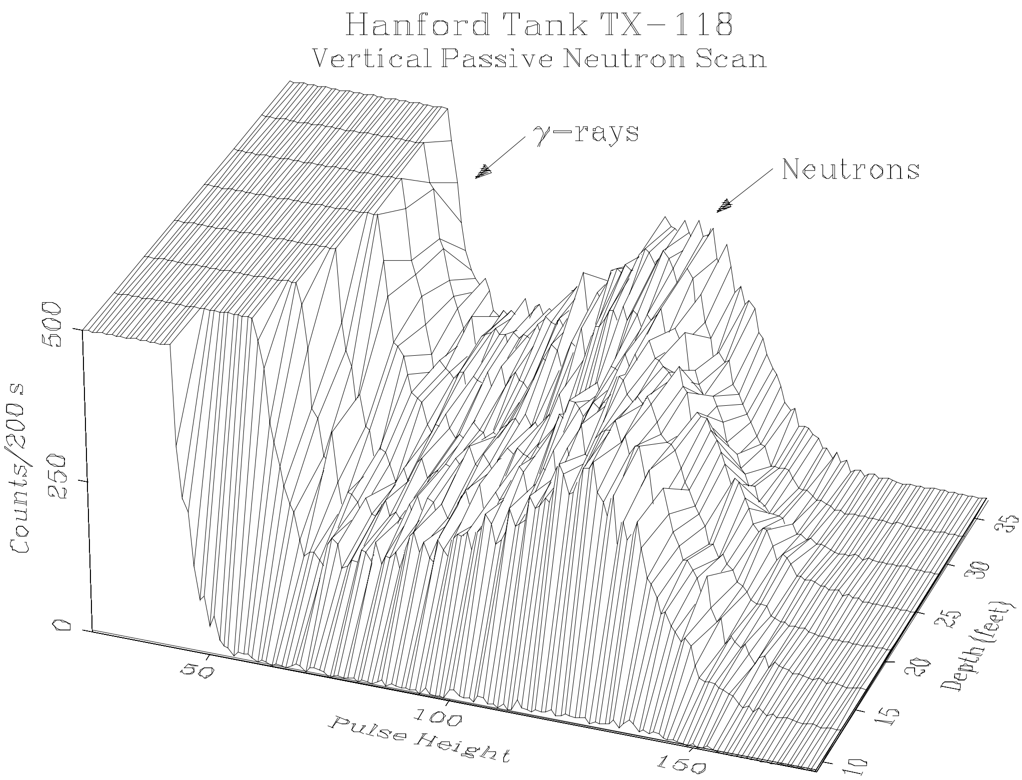

Fig. 3 Counting rate versus pulse height for 8 different depths of detector 3 inside the LOW of tank TX-118 at WHC. The depth is given in feet in this figure and in Fig. 6 so that they can readily be compared to Fig. 1 which was supplied by WHC. The depth as given in this figure is from a fiducial point at the top of the LOW. The depth (d) in this figure can be converted to the "Feet from Bottom of Tank" (f) in Figs. 1 and 6 using the equation; f = 48-d. For example, the first spectrum in this figure was taken at a depth d=9 ft, or equivalently, at f=39 feet from the bottom of the tank.

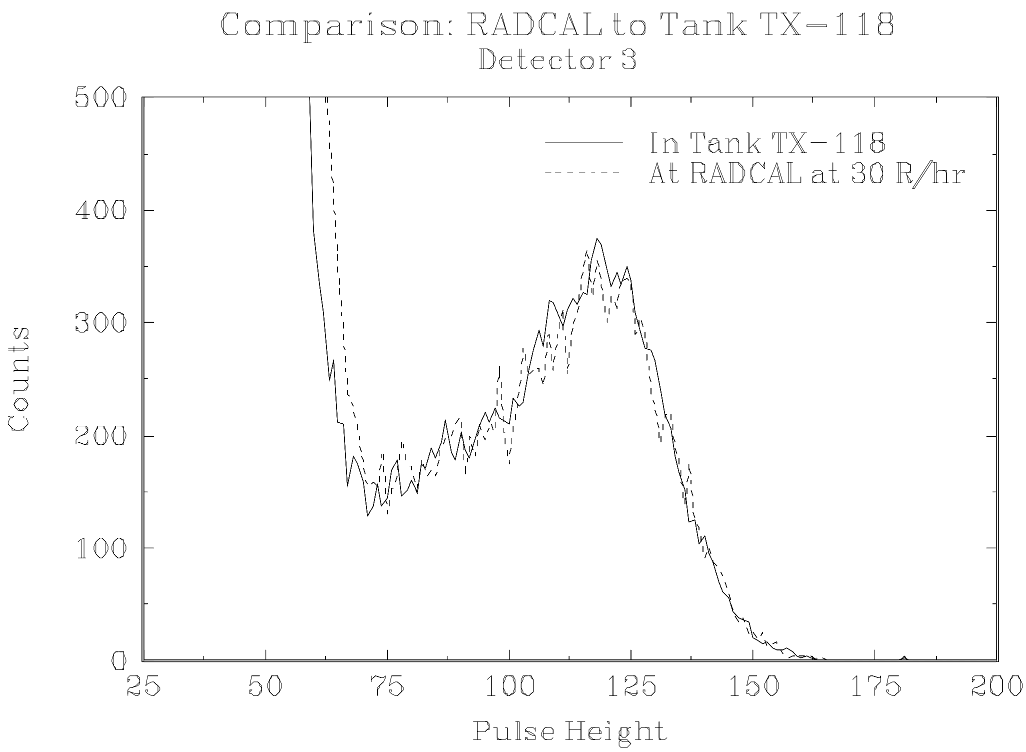

Fig.4. Pulse-height spectra taken with detector 3 at RADCAL at ORNL and in tank TX-118 at WHC. The RADCAL measurements were made at a γ-ray exposure rate of 30 R/hr. Because the RADCAL data were taken with a different gain and a different absolute neutron counting rate (lower than the WHC measurement) they have been scaled to the WHC results.

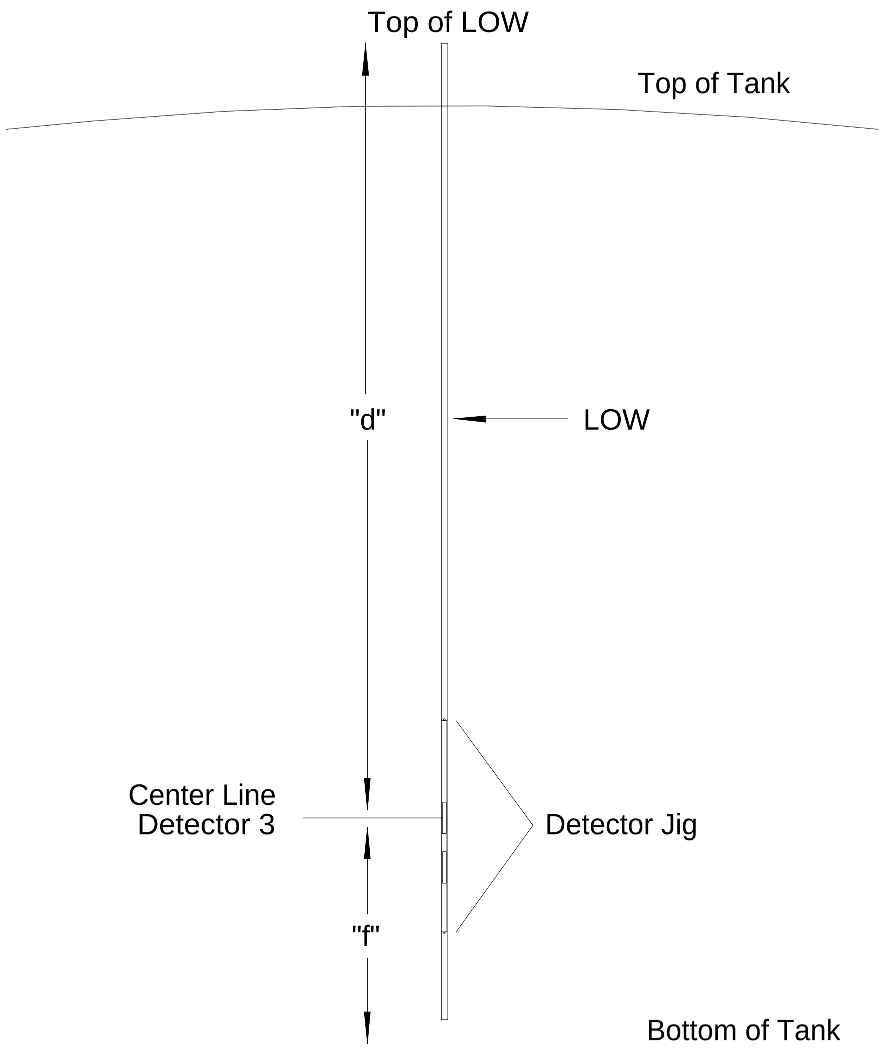

Fig. 5. Schematic diagram of the detector apparatus inside the LOW of tank TX-118 at WHC. The main apparatus was contained in a 3.1-m-long pipe, called the detector jig, which held both detectors, their preamplifiers, and a DC-to-DC converter which supplied high voltage to the detectors from the input low voltage. The distance from the center of the detector to the bottom of the tank, “f”, is used in Figs. 1 and 6. The distance from the top of the LOW riser to the center of the detector, “d”, is used in Fig. 3.

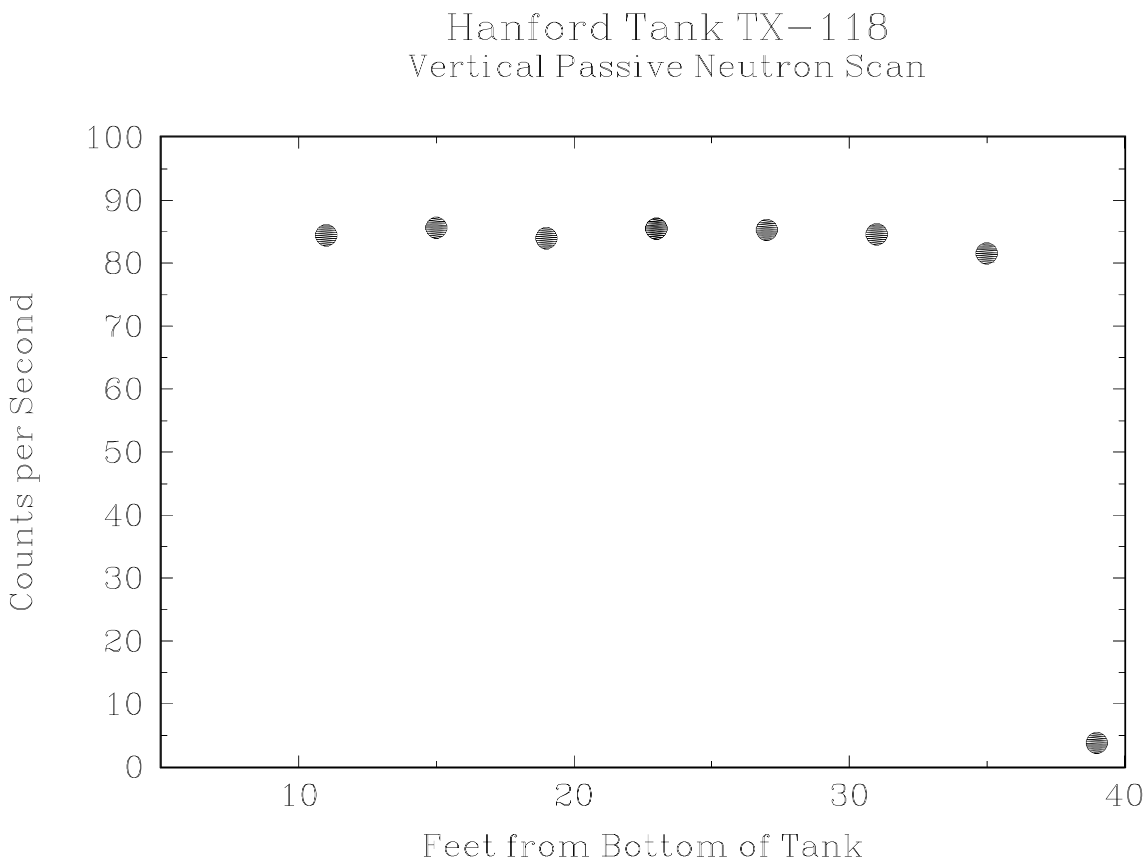

Fig. 6. Counting rate versus the distance of detector 3 from the bottom of tank TX-118 at WHC. The counting rates were calculated by summing the counts above the γ-ray noise for the various spectra in Fig. 3.

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