Radiation Exposure Rate And Liquid Level Measurement Inside A High Level Liquid Waste (HLLW) Storage Tank

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Abstract

An instrument based on an inexpensive, small silicon diode has been developed and used to measure, for the first time, the gamma radiation exposure rate profile inside a 6.4 mm diameter re-entrant thermo-well tube, immersed in the highly radioactive liquid solution in an HLLW storage tank. The measurement agrees with previous calculations of exposure rate, and provides confirmation for safe and effective radiation work plans and material selection for investigations and remediation of the storage tank facility. The measured radiation exposure rate profile is also used to confirm that the position of tank internal structures have not changed because of aging and corrosion, and to obtain, within a few mm, the level of liquid inside the tank.

1. Introduction

Highly radioactive waste solution from medical radioisotope production at Chalk River Laboratories is stored in a High Level Liquid Waste (HLLW) storage tank located inside a shielded, underground vault. Three thermocouples are used to monitor the temperature of the liquid inside the tank at various depths [1]. The thermocouples are installed, inside 6.4 mm (0.25 inch) inner diameter stainless steel re-entrant tubes or thermo-wells that penetrate the tank to different depths. Recently, radioactive liquid from the tank was found to have leaked into the shorter two of the three thermo-wells. This discovery instigated an extensive investigation and corrosion assessment of the thermo-wells and of the storage tank, and the formulation of a strategy to repair the thermo-wells. Various assessments of the gamma radiation exposure rate inside and around the tank were required to formulate safe radiological work plans, and to evaluate the suitability of materials proposed for thermo-well repairs. Confirmation of these assessments using measurements was desirable, although difficult due to the restricted access and high fields involved.

Radiation fields inside the storage tank and the vault had never previously been measured. Gamma radiation exposure rates were expected to be ~10 Gy/h (~1000 Rad/h) inside the storage tank thermo-wells, based on isotopic and shielding calculations. Commercial off-the-shelf instruments that could fit within the 6.4 mm diameter thermo-well and measure such high radiation fields are not available. Therefore a miniature detector system was developed, at short notice, for this application.

The primary measurement objective using this miniature detector system was to confirm the radiation source term of the tank by measuring the gamma radiation field profile inside the third, intact thermo-well. A second objective was to determine the radioactive liquid level from the measured radiation field profile, in order to provide an independent confirmation of this key storage tank operating parameter.

2. Instrument Development

2.1 Sensor

The most common type of sensor used for measuring radiation dose rate is a gas ionisation chamber. These chambers are generally larger than 6.4 mm in diameter. Silicon (Si) p-n junction diodes operating in pulse mode are also used, especially in small, personal dosimeters. The total charge generated by a Si p-n junction diode is well known to be proportional to the ionization energy deposited in the diode depletion region [2]. Since radiation dose is by definition equal to the ionization energy deposited, the total charge is a direct measure of radiation dose. After some experimentation, a small Si p-n diode, operating in current mode was chosen as the sensor for this application. A variety of small, inexpensive Si diodes were readily commercially available because of their extensive use in electronic circuits. Unlike an ionisation chamber, a Si diode does not require high voltage for operation, which considerably simplifies electrical connections and signal amplification. Current-to-voltage amplifiers capable of amplifying the expected $\sim 10^{-10}$ A diode output current for this application are also readily available commercially.

The radiation sensor for this application was selected from a set of commonly available commercial Si diodes, based on size and sensitivity. A set of similar sized diodes was exposed to a radiation exposure rate of 600 Gy/h (60 k Rad/h) inside a Co-60 gamma-cell. The diode with the highest output current, approximately 25 nA, was selected. The selected sensor was a potted Si p-n diode with external potting dimensions 5 mm length and 2 mm diameter, small enough to easily fit within the 6.4 mm inner diameter of the thermo-well.

A separate series of measurements were conducted to assess radiation degradation and temperature sensitivity for a set of Si diode radiation sensors, including the sensor selected for this application. Based on these studies, the change in diode output current caused by radiation degradation and temperature variation was predicted to be negligible over the course of this measurement.

2.2 Probe and readout instrumentation

The starting plan was to deploy the sensor into the thermo-well by pushing a flexible coaxial signal cable attached to the sensor. It was soon discovered that small changes in cable capacitance, caused by flexing the cable, caused transient currents that would overwhelm the expected signal. Therefore this plan for deployment was abandoned, and it was recognised that an almost rigid signal cable connection between sensor and amplifier was required.

Running the signal cable inside a 4.6 m long, 6.35 mm outer diameter stainless steel tube satisfied the above requirement. The probe tube assembly and sensor mounting arrangement is shown in Figure 1. The probe tube was designed to be pushed vertically into the HLLW tank thermo-well, by hand, for radiation field measurements. The tube exterior was marked at 10 cm intervals to facilitate position measurement. The sensor diode was soldered to one end of a low-noise, double-shielded, coaxial signal cable, running inside the probe tube, as shown in the Figure 1. The other end of the signal cable connected to the input of a high-gain current-to-voltage amplifier, attached to a mounting plate welded to the top end of the probe tube. By pulling the amplifier end of the signal cable up by a few mm, the sensor could be retracted inside the probe tube for mechanical protection and partial radiation shielding. Similarly by pushing the amplifier end of the cable down by a few mm, the sensor could be pushed outside the probe tube to obtain an unshielded reading.

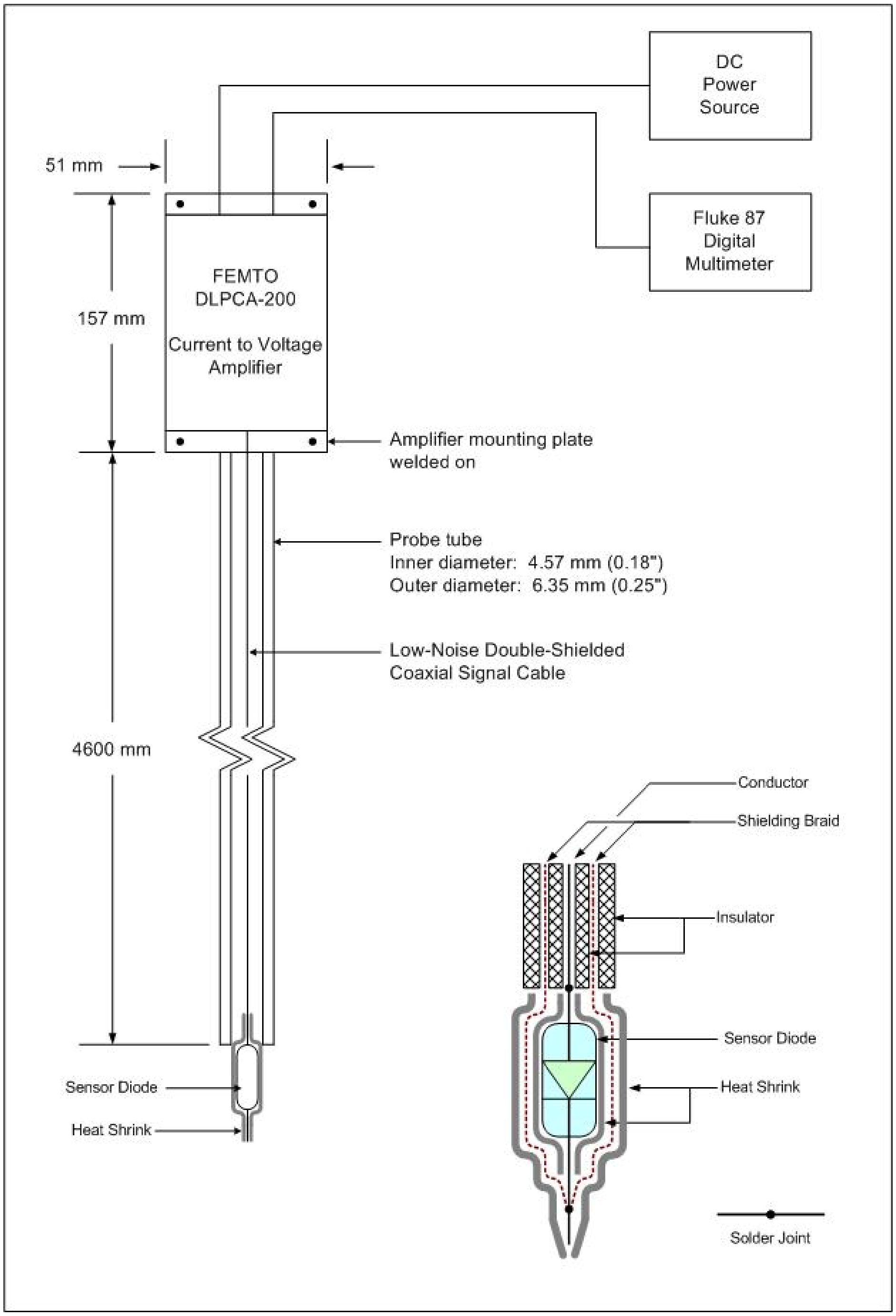

Figure 1 Probe tube assembly and sensor mounting detail.

The current-to-voltage amplifier could be set to any appropriate gain factor ranging from 10^3 V/A to 10^{11} V/A, with an output voltage ranging from 0 to 10 V. The amplifier included a 10 Hz, low-pass filter for rejecting noise. A digital multimeter was used to read out the voltage output of the amplifier. The digital multimeter was set up to sample the voltage signal at 1000 Hz, and to average the sampled signal for 5 s, to further reduce any spurious ac component of the signal.

2.3 Sensitivity and linearity

A source check of the assembled instrument was carried out in an irradiation facility at Chalk River Laboratories, using a calibrated (nominally 30 Ci) point-like ^{137}Cs source. The source-to-sensor distance was varied to obtain gamma radiation exposure rates ranging from approximately 0.6 Gy/h (60 Rad/h) to 3 Gy/h (300 Rad/h), as calculated by the inverse square law. The sensor output current was directly proportional to the exposure rate in this range, as shown in Figure 2. Correcting for amplifier dc offset, this source check gives a sensor output current sensitivity of 0.44 pA (Rad/h)$^{-1}$. The measured sensitivity is consistent with the output current, ~25 nA, obtained at a much higher radiation exposure rate, ~600 Gy/h, demonstrating that the instrument response is linear over a large range of exposure rates, as expected.

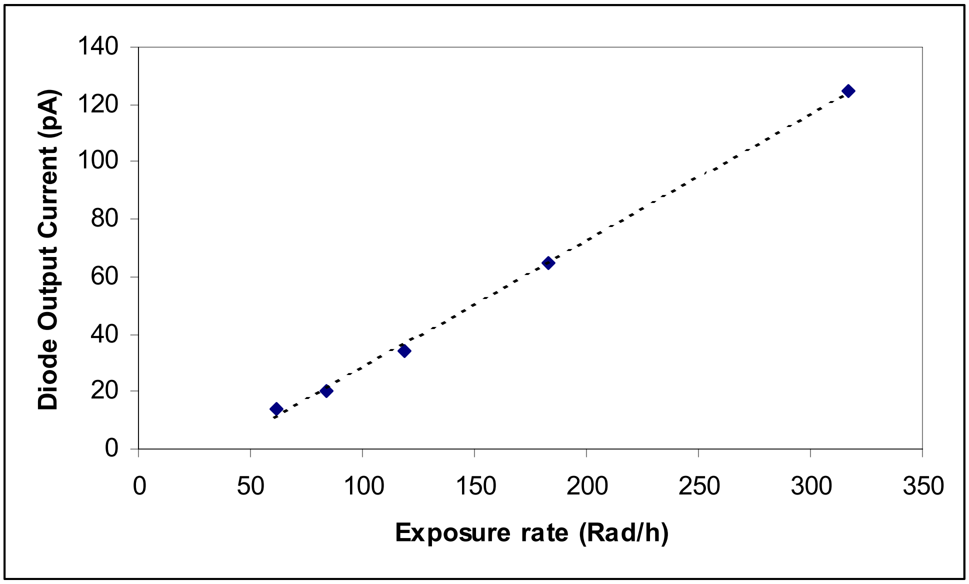

Figure 2 Result of a ^{137}Cs source check in an irradiation facility.

3. Radiation field measurement in HLLW storage tank

3.1 Structural and procedural details

The storage tank is a sealed horizontal stainless-steel cylinder 2743 mm in diameter containing the radioactive liquid. This inner tank is installed inside a cylindrical secondary containment tank 2883 mm in diameter (see Figure 3). Steel plates inside the inner tank support a series of eight horizontal pipe loops, called heating coils 1 to 8, numbered sequentially starting from the bottom of the tank. The heating coils containing pure, deionized water that can be heated or cooled and circulated on demand to control the temperature of the radioactive liquid in the tank.

The tank is in a vault below ground level (reference position 00 at the right of Figure 3). Concrete blocks that form the roof of the vault provide radiation shielding. A conduit in the shielding blocks is used to insert a thermocouple vertically into the thermo-well. A conical “receiving cup” is provided to guide the thermocouple into the 6.4 mm thermo-well bore. The thermo-well tube passes through an opening in the outer tank and is welded to the inner tank. The thermo-well is also constrained near the tank midpoint by a steel support plate ("TW Support" in Figure 3). For the radiation field measurement, the thermocouple was temporarily removed from the thermo-well. The probe with sensor retracted was manually inserted past the receiving cup and in to the top of the thermo-well tube. Once inside the thermo-well, the sensor was pushed out of the probe tube. The probe with exposed sensor was manually inserted down the thermo-well bore, and sensor output readings were taken at predetermined positions in intervals ranging from 20 mm to 100 mm. In the vicinity of the liquid surface, identified by an abrupt change in slope of the radiation profile, readings were taken at 5 mm intervals. When the sensor was close to the bottom of the thermo-well, it was retracted inside the probe tube. Readings were taken at approximately 2 mm intervals during retraction, to gauge the radiation shielding effect of the stainless steel tube.

The probe tube with sensor retracted was then inserted all the way to the bottom of the thermo-well. The measured position of the thermo-well bottom was subsequently used as a datum to determine the position of tank internal structures and of the liquid level with respect to the bottom of the tank.

3.2 Radiation profile measurement

The measured radiation exposure rate is plotted against the distance from the bottom of the thermo-well in Figure 4. Proceeding upwards from the bottom of the thermo-well, i.e., to the right from the 0 mm position in Figure 4, there are several distinct "dips" or local reduction in the exposure rate profile. The positions of these dips coincide with the expected positions of heating coils 2, 3, and 4 (see Figure 3). These sharp local reductions in radiation field are apparently caused by the absence of liquid radioactive source material, combined with the radiation shielding effect of the water and stainless steel in the heating coils. Equally interesting is the lack of dips corresponding to heating coils 5, 6, 7, and 8, and a general reduction of the average exposure rate in the top half of the tank, compared to the bottom half. Lack of liquid radioactive source and shielding caused by water and stainless steel in the vertical inlet-outlet pipes for heating coil 5, running immediately next to the thermo-well as shown in Figure 3, explain these two features.

The large dip in exposure rate near the tank centreline corresponds to the position of the "TW Support" in Figure 3 – a 3 inch wide, ¾ inch thick steel plate – which provides significant shielding from radiation.

An abrupt change in the slope of the exposure rate profile, at about 2140 mm above the thermo-well bottom, marks the interface between the high fields inside the radioactive liquid, and the relatively low fields in the vapour space above the liquid. This feature is discussed in more detail in section 3.3.

The radiation exposure rate decreases rapidly outside the tank walls to a level of approximately 100 Rad/h in the vault above the tank. This measurement agrees with the value obtained by a conventional ionization chamber based instrument lowered into the vault through the shielding block conduit. Finally, as plotted at the extreme right hand side of Figure 4, the exposure rates inside the shielding blocks are essentially below the threshold of detection for this instrument.

Given the variation in exposure rate caused by radiation shielding from various in-tank components as described above, it is judged that the best estimate of exposure rate for comparison with calculations of the radioactive liquid source term is the maximum value measured inside the thermo-well – 11.5 Gy/h (1150 Rad/h). This is in good agreement with the calculated value.

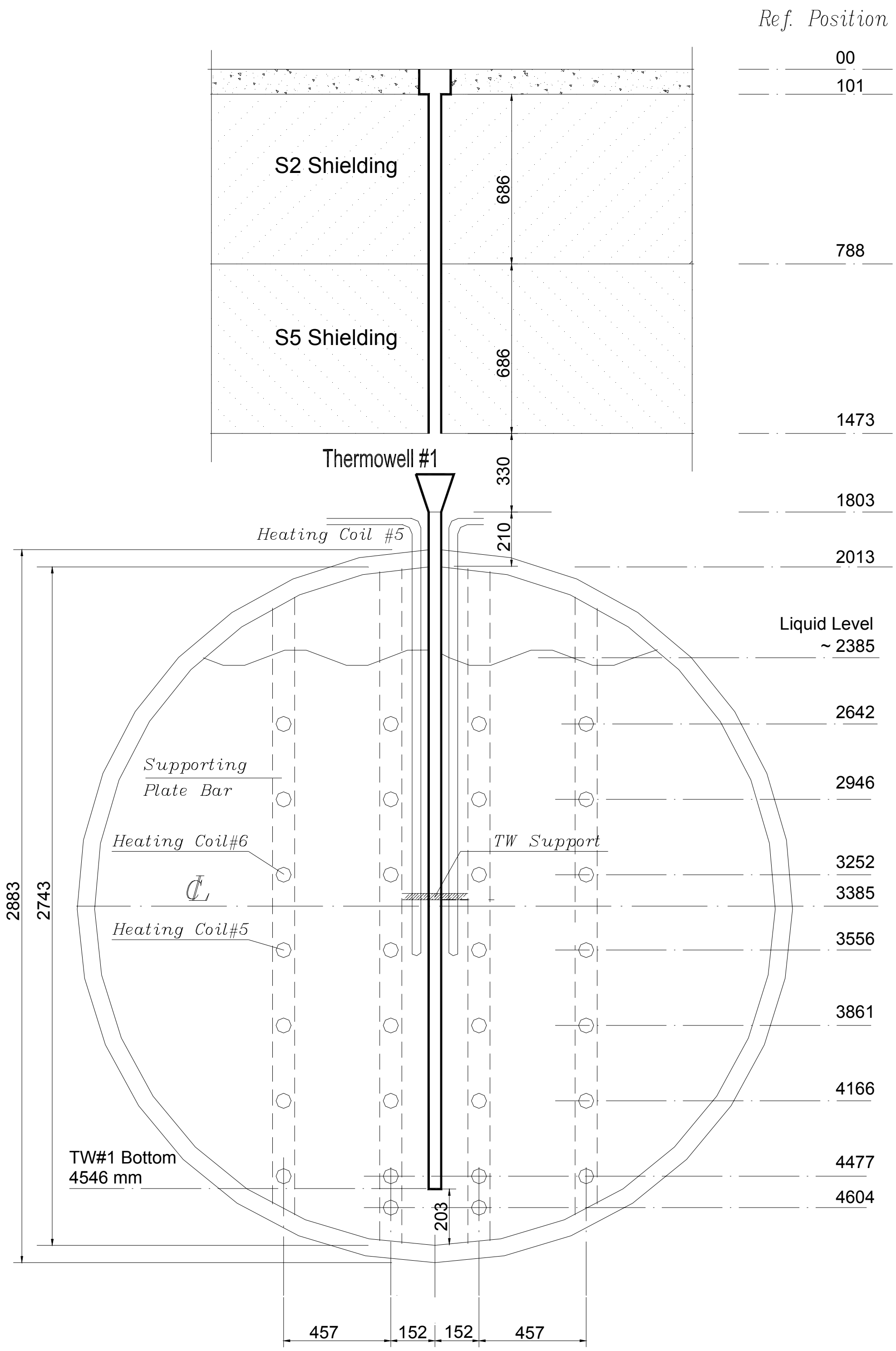

Figure 3 Schematic cross-section of the HLLW storage tank at the thermo-well. All dimensions are in mm. Dimensions on the right hand side refer to nominal distances from the floor level. Please note that the diagram is not to scale.

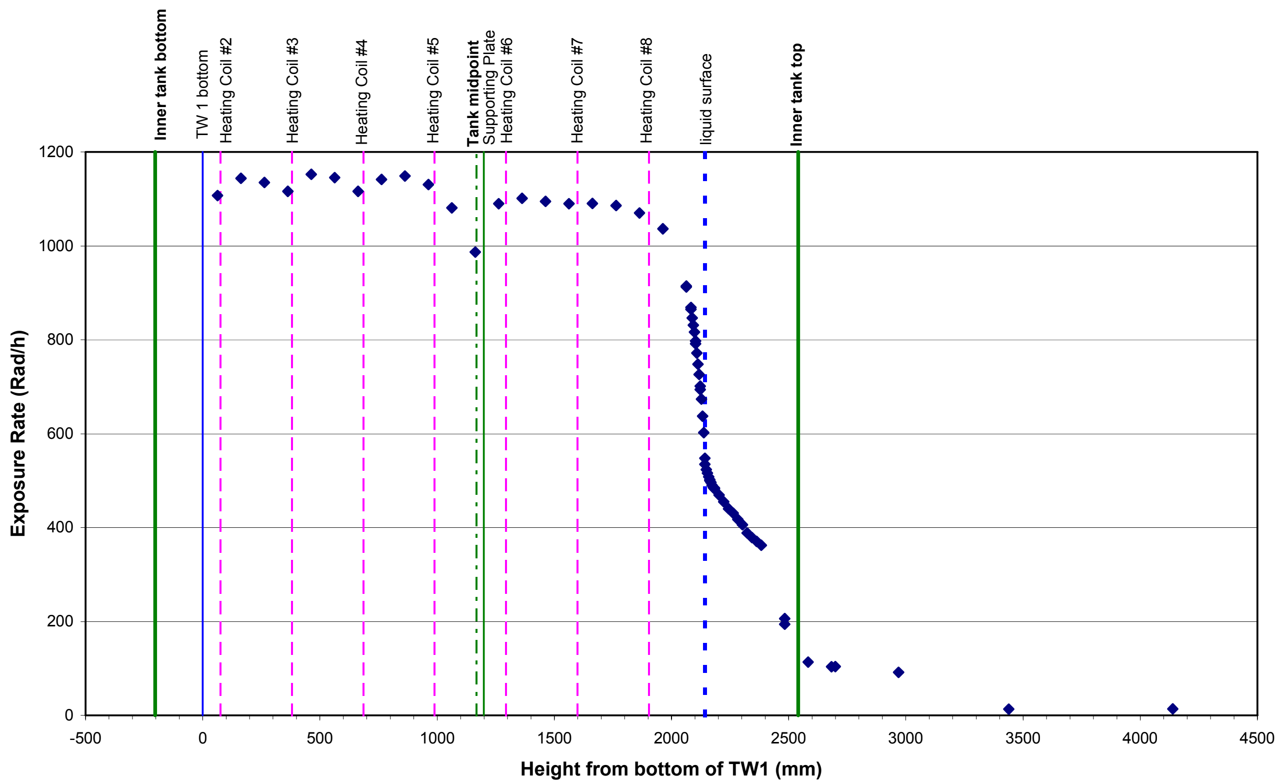

Figure 4 Radiation field profile inside the thermo-well.

3.3 Liquid level

As mentioned in section 3.1, "fine data" measurements at 5 mm intervals were taken around the approximate liquid level, indicated by an abrupt change in slope of "coarse data" taken at 20 mm intervals. The coarse measurements and fine measurements are plotted against height from thermo-well bottom in Figure 5. The abrupt change in slope of the exposure rate profile at 2143 mm above the thermo-well bottom clearly indicates a boundary between two media with very different radioactivity source terms and radiation propagation properties, namely, radioactive liquid and air. It is apparent from Figure 5 that the liquid level is determined within a measurement uncertainty of 5 mm or less.

3.4 Radiation shielding by stainless steel tube

As mentioned in section 3.1, after the radiation profile measurements were completed, and the probe was close to the bottom of the thermo-well, the sensor was retracted inside the probe tube, while taking radiation exposure rate readings at approximately 2 mm intervals. The result, shown in Figure 6 is that the 0.9 mm wall of the stainless steel probe tube causes a 2.7 % reduction in radiation exposure rate when the sensor is completely retracted inside the tube. This information can be used to confirm and extrapolate shielding calculations, taking into account the gamma-ray spectrum, bremsstrahlung x-rays, and decay and build-up factors in the radioactive liquid and the stainless steel thermo-well walls.

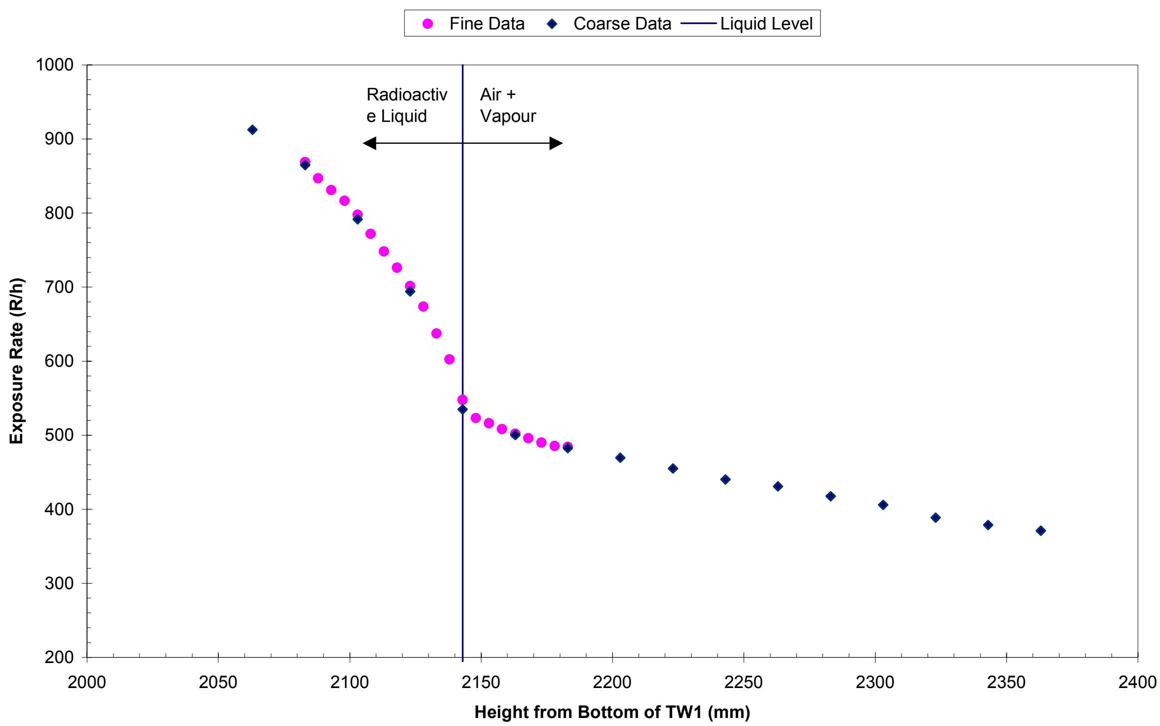

Figure 5 Detailed exposure rate profile near the liquid surface.

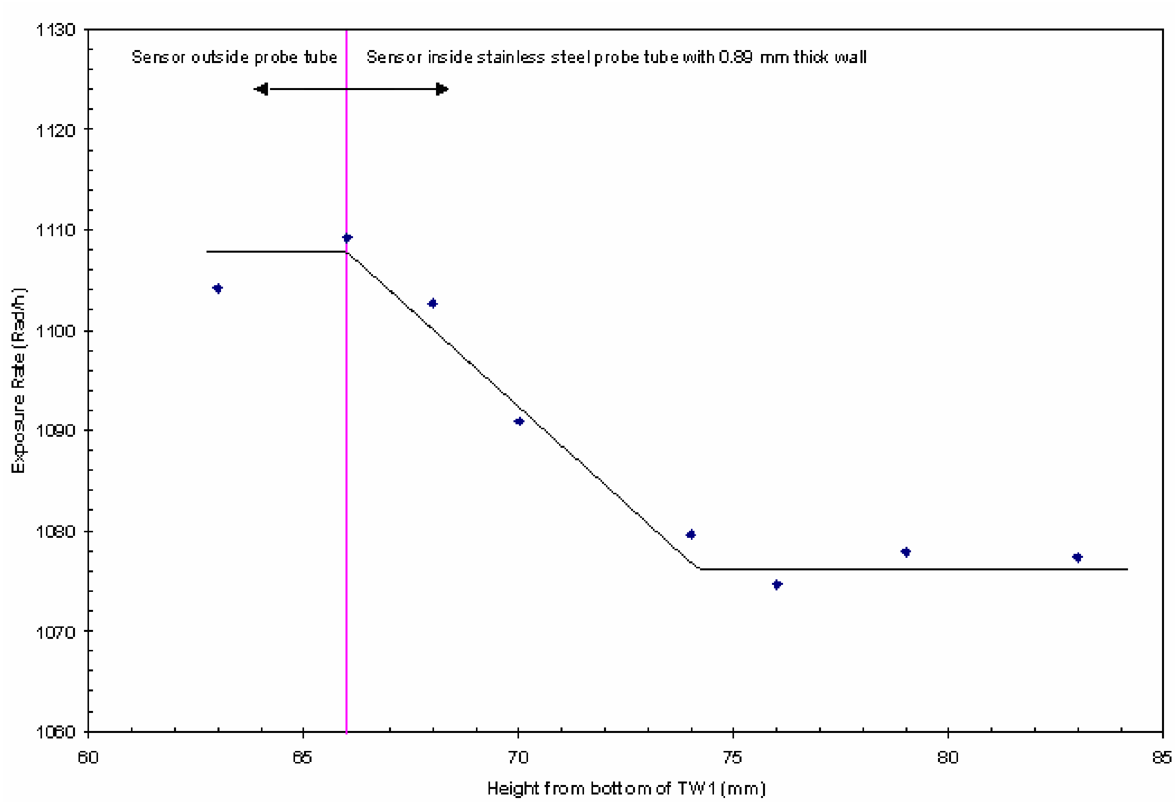

Figure 6 Radiation shielding effect of probe tube wall. A line is drawn through the data points to guide the eye.

4. Conclusions

A detector system was developed and used to measure, for the first time, radiation exposure rates inside an HLLW storage tank containing highly radioactive liquid. The measured exposure rate of 11.5 Gy/h agreed with the value expected from isotopic and shielding calculations. The experimental agreement provided confidence in the results of such calculations for preparing radiological work-plans, and for specifying radiation tolerance of materials, for tank investigation and remediation.

An unexpected result of the radiation exposure rate profile measurement was the detection of in-tank components via their radiation shielding effect. The measurement confirmed that the detected components, particularly heat exchange pipes and support plates, were intact and had not moved after prolonged immersion in the corrosive liquid environment.

The liquid level in the tank was derived from the exposure rate profile measurement to within 5 mm measurement uncertainty. This measurement of liquid level provided a useful independent confirmation of this crucial control parameter required for tank operation.

This work experimentally validates the concept of using an inexpensive, small, commercially available silicon p-n junction diode as the sensor of a detector system for measuring high radiation fields.

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6. References

[1] S. Yue, and B. Sur, "A thermocouple self-heating test for leak detection in a High Level Liquid Waste (HLLW) storage tank", *Proc.* 28th Annual Conference of the Canadian Nuclear Society, 2007.

[2] Knoll, G.F., *Radiation Detection and Measurement*, 2nd Edition, John Wiley & Sons, 1989.